Radiative lifetimes in undoped and *p*-doped InAs/GaAs quantum dots

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We investigate the effect of *p* doping on the luminescence properties of InAs/GaAs self-assembled quantum dots (QDs). Continuous-wave and time-resolved photoluminescence measurements are obtained as a function of temperature and used to extract the radiative lifetime of the QD ground state. We find that the low-temperature luminescence lifetime decreases from $\sim$1200 to $\sim$700 ps for QDs doped with 0 and 10 holes/dot, respectively. The radiative lifetime of the undoped QDs increases monotonically with temperature and is consistent with Boltzmann spreading over dark states. The luminescence intensity from the heavily doped QDs changes much less with temperature compared with the undoped QDs and we attribute this to the presence of holes in ground states at higher temperatures.

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I. INTRODUCTION

In(Ga)As/GaAs quantum dots (QDs) have attracted considerable attention over the last decade for their potential use in a range of optoelectronic devices, including laser diodes, single-photon sources, and quantum information processing applications. The three-dimensional confinement of electrons and holes in the QDs gives rise to discrete electronic states and parity-allowed transitions, which result in atomiclike emission features from individual QDs; emission from QD ensembles is inhomogeneously broadened by variations in QD size and composition. A key feature of the discrete electronic states in QDs is that thermal spreading of carriers over available states is expected to be suppressed, leading to temperature insensitivity of QD devices.[1] Although the behavior of QD devices operating at low temperatures is close to this ideal, toward room temperature there is evidence of thermal spreading of the carrier distribution over the QD states and carrier escape from the QDs.[2–7] As a consequence, QD lasers exhibit disappointingly low critical temperatures of $T_0 \sim 50-100$ K around room temperature,[8,9] comparable with InP-based quantum well (QW) devices. Attempts have been made to increase the room-temperature value of T_0 by *p* doping to reduce the effects of hole escape, which is expected to be more prevalent than electron escape. *p* doping was previously proposed for QW structures[10] but the expected improvements in device performance were not realized due to additional optical loss and nonradiative mechanisms associated with the required high level of *p* doping. However the lower density of states associated with QDs suggests that a relatively low level of *p* doping may be sufficient to obtain improvement in the characteristics of QD devices.[11] Indeed, higher T_0 around room temperature and higher modulation speeds in *p*-doped lasers have since been demonstrated.[12–14] A further reason for studying low to moderately *p*-doped QDs is to understand their role in the performance of QD-based spintronics devices such as QD spin light-emitting diodes, where faster carrier capture times,[15,16] faster carrier relaxation times,[15,17] and longer spin lifetimes[15,18,19] have been reported for *p*-doped structures. *p* doping is thought to increase the spin lifetime in QDs by counteracting the effects of the electron-hole exchange interaction, one of the principal mechanisms of spin relaxation in QDs.[20] The few published reports of fundamental studies of *p*-doped QDs investigated samples having a large doping level[21,22] which masks carrier redistribution effects. In this paper we study both time-resolved and continuous-wave photoluminescence (PL) obtained from undoped and *p*-doped QD structures, covering a range of doping levels up to 10 holes per QD. By combining these data we extract the radiative lifetime of the ground state (GS) over a wide temperature range. Previous studies have presented the luminescence lifetime rather than the underlying radiative lifetime[5,23–26] or have estimated the radiative lifetime only under restricted conditions such as low temperature (for example, from single QD spectroscopy[27]). We then show that the observed temperature-dependent trends may be explained by considering the thermal spreading of carriers over the available QD states and carrier redistribution across the QD ensemble.

II. EXPERIMENTAL DETAILS

The samples were grown by molecular-beam epitaxy on semi-insulating GaAs(100) substrates. Following desorption of the oxide layer, a 200 nm GaAs buffer layer was grown, followed by a 50 nm $Al_{0.3}Ga_{0.7}As$ layer. A further 15 nm of undoped GaAs was grown then the surface was annealed under an As flux at a substrate temperature of 580 °C for 10 min before growth of the QD layer by deposition of 2.4 monolayers (MLs) InAs at a growth rate of 0.018 ML s^{-1} at a substrate temperature of 492 °C. The QDs in the undoped sample were capped by 15 nm GaAs; the doped samples were capped with 13 nm undoped GaAs and followed by 2 nm Be-doped GaAs. A further 50 nm $Al_{0.3}Ga_{0.7}As$ and a 50 nm GaAs cap completed the structure. An atomic force microscopy image obtained from an uncapped QD layer grown under the same conditions is shown in Fig. 1 and indicates a QD density of 2×10^{10} cm^{-2}; this allowed us to determine the Be doping level corresponding to 0, 1, 3, or 10 holes per QD. The AlGaAs blocking layers confine both the photoexcited electron-hole pairs and the doped holes close to the QDs.

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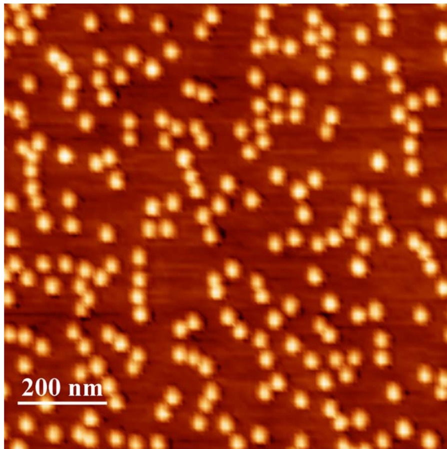

FIG. 1. (Color online) 1 μm×1 μm AFM image obtained from an uncapped QD layer grown under the same conditions as QDs in the capped samples. The average island height in the uncapped layer is 8 nm.

Continuous-wave PL was obtained using a Ti:sapphire laser operating at 790 nm, at excitation densities of 0.5 W cm^{-2} (low power, LP) or 600 W cm^{-2} (high power, HP). The emitted light was dispersed by a 0.85 m double-grating monochromator and detected with a liquid nitrogen-cooled Ge photodiode using standard lock-in techniques. The time-resolved measurements were obtained by exciting the samples with the Ti:sapphire laser operating in pulsed mode (2.4 ps pulses), also at 790 nm. The PL was recorded by a microchannel plate with an extended S1 response, using a time-correlated single-photon counting technique. After deconvolution with the system response, the time resolution was determined to be around 30 ps.

III. RESULTS

Figure 2 shows the continuous-wave PL spectrum obtained from the undoped sample at 12 K under either LP (dashed line) or HP (solid line), normalized to the GS emission peak. Due to the low growth rate used, the QDs are of a highly uniform size and composition and so the inhomogeneously broadened PL emission has a relatively narrow full width at half maximum (FWHM) of 25 meV. A high energy emission tail is observable even under very low power excitation and we attribute this to recombination in a small number of smaller QDs that have GS emission energies roughly coincident with the first excited-state (X1) emission of the majority of larger QDs.[28] However the narrow FWHM allows us to deconvolve the contributions of the GS and X1 emissions even at HP. The inset of Fig. 2 shows PL spectra obtained under similar conditions from the most heavily doped sample (10 holes/QD). Again, GS and X1 emissions are easily distinguishable at HP despite an increase in the PL FWHM to 36 meV, which is consistent with previous reports.[17,29]

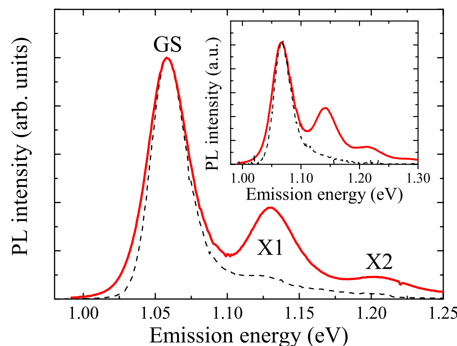

FIG. 2. (Color online) Low-temperature (12 K) PL spectra obtained from undoped sample at LP excitation (0.5 W cm^{-2}, black dashed line) and HP excitation (600 W cm^{-2}, red/gray solid line). Inset: 12 K PL spectra obtained from the most heavily doped sample (10 holes/QD) with LP excitation (black dashed line) and HP excitation (red/gray solid line). The spectra have been normalized to the GS emission peak.

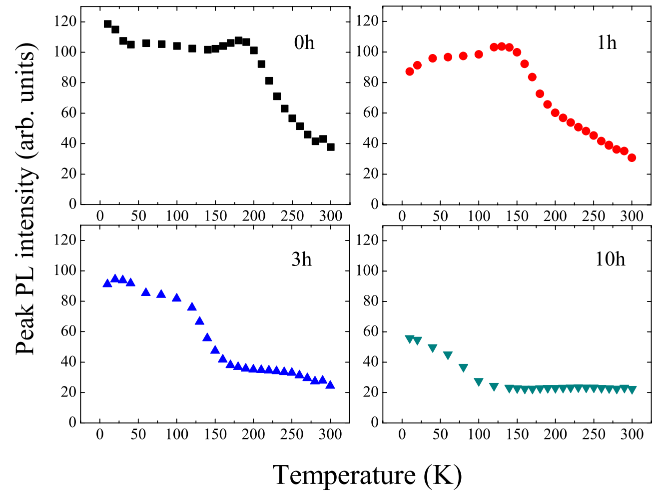

FIG. 3. (Color online) Temperature dependence of the peak PL intensity of the GS emission for the four samples, obtained under HP excitation conditions.

Figure 3 shows the peak GS emission intensity obtained under HP excitation for all four samples as a function of temperature. These data are presented on a linear scale rather than as Arrhenius plots for ease of interpretation. For the undoped sample, GS emission is reasonably constant up to 190 K and then falls by a factor of ~3 by room temperature. With increasing doping level, the low-temperature PL intensity is reduced; this has previously been attributed to an increased density of dopant-related trap states in the *p*-doped GaAs.[29] Also, as the doping increases, the temperature at which the luminescence starts to quench tends to lower temperatures and is not clearly identifiable for the most heavily doped sample. At higher temperatures, particularly for the samples doped with 3 or 10 holes per QD, the PL emission intensity reaches another plateau at a temperature which decreases with increasing doping. Although the low-temperature GS peak PL intensity is reduced for samples with increased doping, the absolute reduction in PL with increasing temperature is less pronounced. It is usual to see reductions in the PL intensity over this temperature range by at least an order of magnitude[3–5] rather than the factors of 2–3 seen for these samples and we attribute this to the high

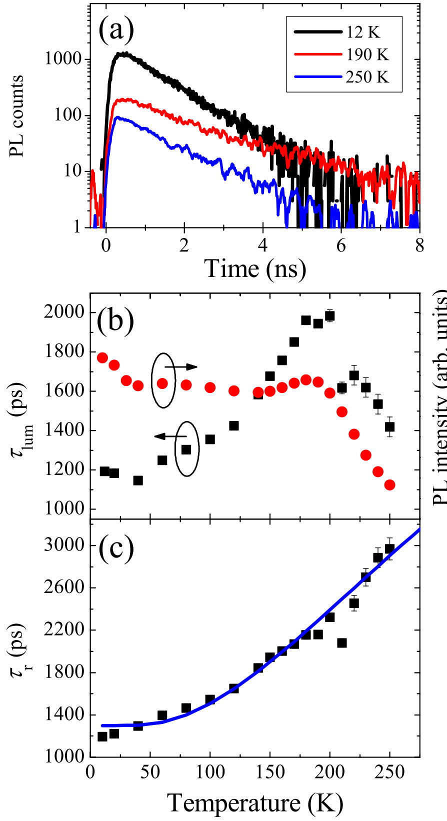

FIG. 4. (Color online) (a) Time-resolved PL data measured at the GS peak emission energy and obtained at high excitation power (600 W cm^{-2}) from the undoped sample at 12 K (black), 190 K (red/light gray), and 250 K (blue/dark gray). (b) Temperature dependence of GS luminescence lifetime (black squares) and continuous-wave peak GS PL intensity (red/light gray circles) obtained under high power excitation. (c) Temperature dependence of GS radiative lifetime (black squares), extracted from time-resolved and continuous-wave PL data presented in (b), with model fit (blue/dark gray solid line).

excitation conditions and the presence of the AlGaAs layers, which prevent diffusion of escaped carriers away from the QDs.

Figure 4(a) shows representative time-resolved PL decay curves corresponding to peak GS emission from the undoped sample at 12, 190, and 250 K. The PL data are deconvolved from the system response and are well fitted by single exponentials at all temperatures. Figure 4(b) shows GS luminescence lifetime, τ_{lum}, obtained from single exponential fits of the time-resolved PL data measured at the GS peak from the undoped sample (squares). Also plotted is the peak GS PL intensity obtained from the continuous-wave spectra (circles), previously shown in Fig. 3. Due to the limited response of the microchannel plate at long wavelengths and the redshift of the QD emission with increasing temperature, our time-resolved PL measurements are limited to a maximum temperature of 250 K. The luminescence lifetime gradually increases from a low-temperature value of 1200 ps to a maximum of 2000 ps at 200 K. At higher temperatures the luminescence lifetime then falls, reaching 1400 ps at 250 K. The high-temperature behavior closely matches that of the continuous-wave PL intensity, which exhibits a maximum at ~180 K. A similar temperature dependence of the luminescence lifetime has been observed in previous studies.[5,23–26]

Miller *et al.*[30] have shown that it is possible to extract the radiative lifetime from the time-resolved and continuous-wave data. The luminescence lifetime τ_{lum}, obtained from the time-resolved data, consists of radiative (τ_r) and nonradiative (τ_{nr}) components, such that

$$\frac{1}{\tau_{lum}} = \frac{1}{\tau_r} + \frac{1}{\tau_{nr}}. \quad (1)$$

If carriers are photogenerated at a rate G, then the rate of emission can be written as

$$\frac{dN}{dt} = G - \frac{N}{\tau_{lum}}, \quad (2)$$

where N is the number of excitons. In the steady state, $N = G\tau_{lum}$ and a simple integration yields

$$N(t) = N(0)\exp\left(-\frac{t}{\tau_{lum}}\right). \quad (3)$$

The continuous-wave PL from the ground state, PL_{GS}, can be expressed by

$$PL_{GS} \propto \frac{N}{\tau_r} = G\frac{\tau_{lum}}{\tau_r} \quad (4)$$

and

$$\tau_r \propto \frac{\tau_{lum}}{PL_{GS}}. \quad (5)$$

Thus using the measured luminescence lifetime of the GS emission, together with the continuous-wave PL intensity, a quantity proportional to the radiative lifetime can be extracted across the temperature range considered here. Usually it is assumed that nonradiative processes are negligible at low temperature ($\tau_{lum} \sim \tau_r$), so the value for the GS radiative lifetime at 12 K can be used to scale the radiative lifetimes at higher temperatures. As can be seen from the PL spectrum obtained under HP conditions shown in Fig. 2, the PL intensity from X1 is considerably less than that from the GS and previous measurements have shown that the luminescence lifetime is independent of excitation power up to 600 W cm^{-2},[28] so saturation of PL_{GS} and the contribution to dN/dt by excited-state emission is not expected to significantly affect the analysis in Eqs. (1)–(5). Since trap states due to the doping in the GaAs would only affect the generation and capture of carriers into the QDs, they would not influence the measured decay time τ_{lum} and at higher temperatures any nonradiative processes affecting emission from the QDs will influence both the continuous-wave PL and τ_{lum} so, following the analysis of Miller *et al.*, the actual origin of any nonradiative loss does not affect the determination of τ_r. Figure 4(c) shows the GS radiative lifetime obtained using this method over a temperature range of 12–250 K (black squares). We observe a monotonic increase with temperature across the whole temperature range. To describe this behavior, we adapt a model from Gurioli *et al.*[31] The rate at which

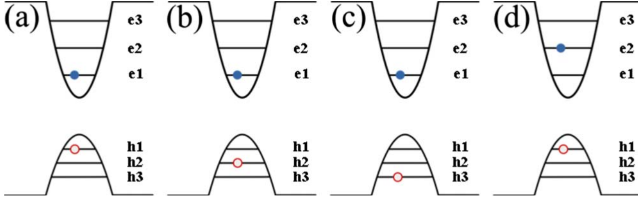

FIG. 5. (Color online) Electron and hole occupancy of states for the four lowest energy configurations of a single electron-hole pair in model QD.

excitons recombine in the GS is given by $\Gamma(T)$ (and by Γ_0 in the limit of low temperature). Figure 5 shows the lowest energy configurations of an electron-hole pair in a model QD with electron and hole energy-level separations of 50 and 20 meV, respectively. Here we consider heavy holes only: in a strongly confined system such as QDs the degeneracy of heavy and light hole states is lifted and previous experimental studies and $\mathbf{k}\cdot\mathbf{p}$ modeling of hole states in InAs/GaAs QDs suggest that they have mainly heavy-hole character.[32–36] If present, light hole states would be expected to be occupied in the most heavily p-doped sample and e1-lh1 optical transitions should be observed even at LP. The single PL peak observed under these conditions (inset of Fig. 2) shows that this is not the case, justifying our premise. To calculate the total radiative rate, Γ, we average over the accessible energy states[37,38]

$$\Gamma = \frac{1}{\tau_r} = \sum_i \Gamma_i p_i, \tag{6}$$

where Γ_i is the radiative rate for the ith state and p_i is the probability of occupation of state i, determined by the partition function, Z,

$$p_i = \frac{g_i e^{-E_i/k_BT}}{Z}, \tag{7}$$

where g_i is the degeneracy of the ith state. Setting the zero energy corresponding to the GS ($i=0$) state then

$$Z = \sum_i g_i e^{-E_i/k_BT} = g_0 + g_1 e^{-\Delta E_h/k_BT} + g_2 e^{-2\Delta E_h/k_BT} + \cdots . \tag{8}$$

If the electron and hole are both in the GS [e1, h1, as shown in Fig. 5(a)], recombination is allowed and occurs at a rate Γ_0. This is the case at low temperature. At higher temperatures, carriers can occupy higher-lying energy levels, with (in order of increasing energy) the hole in h2 [Fig. 5(b)], h3 [Fig. 5(c)], and the electron in e2 [Fig. 5(d)]. Recombination between these energy levels is forbidden, so the recombination rate is zero. This gives

$$\Gamma = \frac{g_0\Gamma_0}{Z} = \frac{\Gamma_0}{1 + \frac{g_1}{g_0}e^{-\Delta E_h/k_BT} + \frac{g_2}{g_0}e^{-2\Delta E_h/k_BT} + \cdots}. \tag{9}$$

Taking the degeneracies of the first, second, and third states to be 2, 4, and 6, including spin degeneracy, then the radiative lifetime of the states is given by

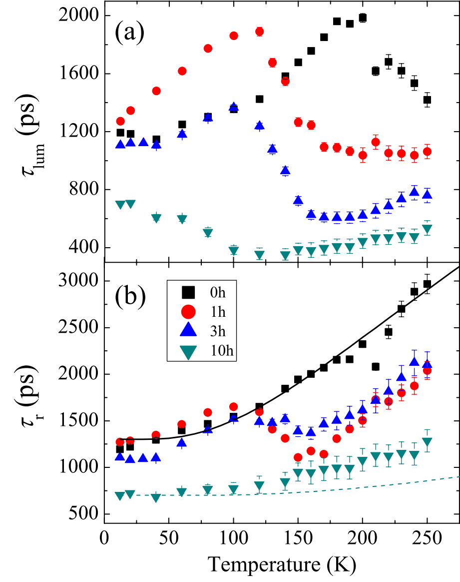

FIG. 6. (Color online) Temperature dependence of (a) GS luminescence lifetime, obtained under HP excitation conditions, (b) GS radiative lifetime for all samples, with fits from the model considering a single electron-hole pair in the QD (black solid line) or full occupancy of the hole states (cyan/gray dashed line).

$$\tau = \tau_0(1 + 2e^{-\Delta E_h/k_BT} + 3e^{-2\Delta E_h/k_BT} + 2e^{-\Delta E_e/k_BT} + \cdots). \tag{10}$$

This equation contains only two parameters, τ_0 and the energy-level spacing. The total state separation is $\Delta E_e + \Delta E_h = 73$ meV, as measured from the continuous-wave PL spectra. Adopting a value for the low-temperature GS radiative lifetime $\tau_0 = 1300$ ps and a hole energy separation of 23 meV, chosen by assuming an electron energy-level spacing of 50 meV, consistent with double resonant spectroscopy on similar QDs,[39] far-infrared spectroscopy on charged InAs/GaAs QDs (Ref. 40) and Coulomb blockade measurements,[41,42] then the temperature dependence of the radiative lifetime from the model is shown as the solid line in Fig. 4(c). There is excellent agreement with the experimental results over the entire temperature range.

The analysis can be extended to the doped samples. Figure 6(a) shows the temperature dependence of the luminescence lifetime for all four samples. Generally, the luminescence lifetime is reduced as doping is increased and for each sample we observe a peak in the luminescence lifetime, which shifts to lower temperatures as the doping level is increased, similarly to the behavior seen for continuous-wave data (Fig. 3). Using the continuous-wave data presented in Fig. 3, the GS radiative lifetime can be extracted for all samples from 12 to 250 K, shown in Fig. 6(b). It is noted that at low temperature, the GS radiative lifetime for the undoped sample and the lightly doped samples (1 hole/QD and 3 holes/QD) are approximately the same but the radiative lifetime for the most heavily doped sample is considerably shorter. This is consistent with previous PL studies[22,29,43] and measurements of an increased radiative recombination rate in p-doped lasers.[44] As the temperature is

increased, the rate of increase in the radiative lifetime for the 10 holes/QD sample is much less than for the undoped sample, as might be expected if the effect of thermal spreading of carriers across the available energy states is countered by the presence of the additional holes.

The previous model can now be extended to the heavily p-doped sample. At low temperature, the increase in lifetime is now associated only with spreading over the electron states. As before, the rate can be calculated by averaging over the accessible energy states,

$$\Gamma = \frac{g_0\Gamma_0}{Z} = \frac{\Gamma_0}{1 + \frac{g_1}{g_0}e^{-\Delta E_e/k_B T} + \frac{g_2}{g_0}e^{-2\Delta E_e/k_B T} + \cdots}. \quad (11)$$

The radiative lifetime of the ground state is now given by

$$\tau = \tau_0(1 + 2e^{-\Delta E_e/k_B T} + 3e^{-2\Delta E_e/k_B T} + \cdots). \quad (12)$$

Equation (12) is plotted in Fig. 6(b) as a dashed line with $\Delta E_e = 50$ meV and $\tau_0 = 700$ ps. The model and deduced lifetimes agree reasonably well below 100 K but the model predicts a smaller rate of increase above this value. A possible explanation is that holes escaping from the charged QDs above 150 K contribute to the increase in τ_r.

The fit to the radiative lifetimes of the undoped sample in Fig. 6(b) [and previously shown in Fig. 4(c)] considers thermal spreading of a single electron-hole pair over the available QD states. In this case the increase in radiative lifetime is mainly determined by the thermalization of the hole. For the most heavily doped case, it is assumed that all the hole states are filled and the temperature dependence of the radiative lifetime is only determined by thermal spreading of the electron. This is a good approximation at low temperatures but at higher temperatures (>150 K), when holes will escape from the QDs, we would expect a deviation from this trend and this is what we see. The two fits provide the limiting cases for carrier dynamics determined by either electrons or holes. A more accurate model of state filling in heavily doped QDs would include nonsequential state filling and the violation of the Aufbau principle for holes in QDs.[42,45,46]

A more complicated behavior is seen for the samples doped with 1 and 3 holes but the values lie within the limits defined by the two models and the experimental data for the undoped sample and the sample doped with 10 holes/QD. We attribute the unexpected reduction in the GS radiative lifetime between 100 and 150 K to the onset of independent hole escape from the QDs.[6,7] In this temperature range, where hole escape can occur but electron escape does not, holes can be redistributed through the QD ensemble, finding QDs containing electrons and so leading to an increase in the radiative recombination rate compared to when carrier occupation of QDs is determined by random population. Hole escape is likely to have an insignificant effect on the undoped sample since there are no excess holes and the AlGaAs barriers will ensure rapid recapture. For the heavily p-doped sample escape of a few holes will again have little effect on the radiative lifetime which is determined mainly by the sizable remaining hole population. Further work is required to confirm this suggestion.

IV. CONCLUSIONS

We have measured the temperature dependence of PL emission obtained from undoped and p-doped InAs/GaAs QD ensembles using both continuous-wave and time-resolved measurements, and have extracted the GS radiative lifetimes for these samples over a temperature range of 12–250 K. We find that the radiative lifetime of emission from QDs increases with temperature much more gradually than for QWs, as is expected due to the in-plane confinement and the increase can be well explained by Boltzmann spreading of carriers over the available states. For heavily doped QDs, the GS radiative lifetime increases more gradually with temperature since the effect of thermal spreading of carriers is mitigated by the presence of additional holes.

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