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## Impact of Lifetime Control on the Threshold of Quantum Dot Lasers

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Despite significant improvements in their properties as emitters, colloidal quantum dots have not had much success in emerging as suitable materials for laser applications. Gain in most colloidal systems is short lived, and needs to compete with biexcitonic decay. This has necessitated the use of short pulsed lasers to pump quantum dots to thresholds needed for amplified spontaneous emission or lasing. Continuous wave pumping of gain that is possible in some inorganic phosphors has therefore remained a very distant possibility for quantum dots. Here, we demonstrate that trilayerheterostructures could provide optimal conditions for demonstration of continuous wave lasing in colloidal materials. The design considerations for these materials are discussed in terms of a kinetic model. The electronic structure of the proposed dot architectures is modeled within effective mass theory.

### Introduction

With continuous progress in colloidal chemistry, semiconductor quantum dots<sup>1</sup>(QDs) have emerged as materials of choice for a variety of optical<sup>2-5</sup> and optoelectronic applications<sup>6, 7</sup>. While QDs rival and even surpass the properties of organic and inorganic phosphors in many ways, the gain performance of these materials is still not optimal<sup>8, 9</sup>. Demonstrations of gain in these materials involve pumping them with high intensity light sources, typically short pulsed lasers. Practical applications therefore demand the development of QDs with significantly improved gain behavior. Here, gain behavior is described using a kinetic model. This provides insights into the critical properties that an ideal gain medium should possess. We then use effective mass theory to demonstrate that suitably designed QDs could act as near ideal gain media. We show that QDs could possibly be modified to exhibit gain under continuous wave (CW) pumping.

### Model

We begin our discussion by considering a highly simplified picture of a gain medium comprising of QDs illuminated by a continuous wave light source. The number of photons absorbed per QD per second is  $f\sigma_A$ , where,  $f$  is the fluence in photons per  $\text{cm}^2$  per second that is provided by the pump

source and  $\sigma_A$  is the optical cross section in  $\text{cm}^2$ . We consider the situation at steady state. Let  $N$  be the probability of a dot to have one exciton and  $B$  be the probability for a dot to have a biexciton. In a typical II-VI QD, the band edge is two-fold degenerate. A single exciton is thus optically transparent while a biexciton can induce gain. A non-excited QD can absorb light at the band edge and therefore induces losses. In this highly simplified picture, the probability of encountering an unexcited QD is  $1 - N - B$ . The gain medium would become optically transparent when this probability equals the probability of finding a biexciton, i.e.  $1 - N - B = B$  or  $1 - N - 2B = 0$ . At threshold, the QD ensemble consists of unoccupied QDs, as well as QDs with single and multiple excitons. In a II-VI semiconductor, a multi-exciton produces the same gain as a biexciton because of the two-fold band edge degeneracy. We therefore restrict the counting of multiexcitons to biexcitons only and neglect the presence of QDs with more than two excitons. This is discussed in greater detail in the supplementary section.

At steady state, the pump source acts on unexcited QDs to produce single excitons with a rate  $\sigma_A(1 - N - B)$ , and on single excitons to produce biexcitons with a rate  $Nf\sigma_A$ . In these expressions  $\sigma_A$  represents the absorption cross section at the pump wavelength. Spontaneous decay of single excitons that occurs with a lifetime  $\tau$  and biexciton decay that occurs with a lifetime  $\tau_B$  constitute loss mechanisms that de-excite singly and doubly excited QDs. The rate equations describing such a medium may therefore be written as:

$$\frac{dN}{dt} = f\sigma_A(1 - N - B) - Nf\sigma_A + \frac{B}{\tau_B} - \frac{N}{\tau} \quad (1)$$

$$\frac{dB}{dt} = Nf\sigma_A - \frac{B}{\tau_B} \quad (2)$$

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These equations imply that single excitons are created by pumping un-excited QDs and are themselves converted to biexcitons upon absorption of light from the pump source. Single excitons can also be created by biexcitonic decay and also annihilated by spontaneous decay. In turn, biexcitons are produced by the absorption of light by single excitons and destroyed by biexcitonic decay. As discussed previously, the conversion of a biexciton into a multiexciton does not affect its gain characteristics. We therefore ignore the changes in biexcitonic populations due to multiexciton formation. At steady state, all probabilities are constant, and therefore the left side of both equations (1) and (2) may be set to zero. By substituting the transparency condition  $1 - N - 2B = 0$  into the resultant equations, one may show that the fluence needed to cause transparency at steady state is given by

$$f(\text{transparency}) = \left( \sigma_A \sqrt{\tau \tau_B} \right)^{-1}$$

This expression suggests the influence of single exciton and biexciton lifetimes on the fluence required for attaining optical transparency. It is helpful to analyze this expression using parameters observed in typical QDs. A 10 nm CdSe QD has an emission lifetime  $\tau$  of  $10 \text{ ns}^{10}$ , a cross section of  $2 \times 10^{-14} \text{ cm}^2$  at 400 nm and a biexciton lifetime  $\tau_B$  of  $700 \text{ ps}^{11,12}$ . Transparency thus occurs at  $f = 10 \text{ kW/cm}^2$  under CW excitation, a fluence that is too high to be practical. Suppose the QDs are somehow modified so that the spontaneous emission lifetime of the QD increases to  $10 \mu\text{s}$ . The onset of gain now occurs at a fluence of  $f = 290 \text{ W/cm}^2$ . This fluence could possibly be achieved by concentrating light from commonly available light sources. This entire discussion is extremely simplified and neglects the role of losses in the lasing process. Despite its limitations, this model nevertheless shows the importance of a long spontaneous lifetime in ensuring gain in a colloidal system.

Motivated by the above discussion, we proceed to construct a more accurate model of a QD laser (see figure 1a,b). The equations governing population and flux inside a QD laser<sup>13</sup> are written as:

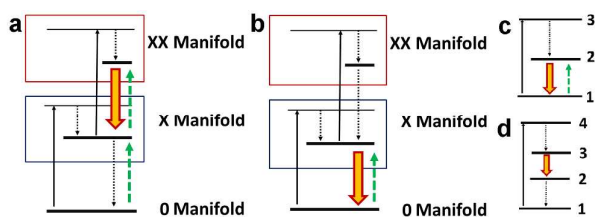
$$\frac{dN}{dt} = W_p(1 - N - B) - W_p N + 2c\sigma\phi(1 - N - B) - 2Nc\sigma\phi + 2Bc\sigma\phi + \frac{B}{\tau_B} - \frac{N}{\tau} \quad (3)$$

$$\frac{dB}{dt} = W_p N + c\sigma\phi(N - 2B) - \frac{B}{\tau_B} \quad (4)$$

Here  $W_p = f\sigma_A$  is the pump rate in units of  $\text{s}^{-1}$ ,  $c$  is the speed of light in the cavity in  $\text{ms}^{-1}$ .  $\phi$  is the photon density in units of photons per  $\text{cm}^3$ .  $\sigma$  is the cross section of stimulated emission that is related to the spontaneous lifetime by

$$\sigma = \frac{\lambda^2}{4\pi n_0^2 \Delta\nu \tau} \left( \frac{\ln 2}{\pi} \right)^{1/2}$$

for a Gaussian line centered at  $\lambda$  with band width  $\Delta\nu$  in a medium with refractive index  $n_0$ . It is important to note that  $\sigma$  is the cross section "per electronic transition". For an unexcited II-VI QD, there are two possible transitions that could possibly cause it to turn into a singly excited QD. A degeneracy factor of 2 thus needs to be introduced when considering stimulated processes by unexcited or doubly excited particles. These equations contain additional  $\phi$  dependent terms that are absent in equations (1) and (2). Equation (3) contains  $2c\sigma\phi(1 - N - B)$  that represents the excitation of an unexcited QD into a singly excited QD due to absorption of band edge light;  $-2Nc\sigma\phi$  represents the conversion of a singly excited QD into an unexcited or a doubly excited QD through stimulated emission or absorption respectively and  $2Bc\sigma\phi$  represents the conversion of a biexciton into a single exciton by stimulated emission. Similar  $\phi$  dependent terms occur in equation (4).



**Figure 1.** Energy level scheme for **a.** QDs with biexcitonic gain **b.** QDs with single excitonic gain **c.** a three level system and **d.** a four level system. Solid black arrows represent the action of the pump. Dotted black arrows represent miscellaneous relaxation pathways, dashed green arrows represent absorption of band edge light, while the thick yellow arrow represents the laser emission.

Physically, the key distinction between equations (1), (2) and (3), (4) is that the latter allow for stimulated processes where QDs interact with the photon density  $\phi$  at the band edge. The photon density  $\phi$  is given by

$$\frac{d\phi}{dt} = 2c\sigma\phi x B - 2c\sigma\phi x(1 - N - B) - \frac{\phi}{\tau_C} \quad (5)$$

This equation accounts for increases in the flux inside the cavity due to biexcitons as well as the removal of photons due to ground state absorption. The photon flux within the cavity can also decay because of losses at various interfaces within the cavity as well as because of the extraction of a laser beam. These processes collectively give rise to a characteristic persistence time  $\tau_C$  of photons inside the cavity. We are

primarily interested in the threshold of a QD laser. At threshold, stimulated emission still does not become significant enough to change the populations appreciably. We therefore neglect terms corresponding to stimulated emission in equations (3) and (4) while setting up the conditions for steady state operation at threshold

$$\frac{d\phi}{dt} = \frac{dN}{dt} = \frac{dB}{dt} = 0 \quad (6)$$

This leads to

$$W_p(1-N-B) - W_p N + \frac{B}{\tau_B} - \frac{N}{\tau} = 0 \quad (7)$$

$$W_p N - \frac{B}{\tau_B} = 0 \rightarrow B = W_p N \tau_B \quad (8)$$

$$2c\sigma x B - 2c\sigma x(1-N-B) - \frac{1}{\tau_C} = 0 \quad (9)$$

$$\rightarrow N = \frac{1+\zeta}{1+2W_p\tau_B}$$

Where for convenience we have defined

$$\alpha = xc\sigma\tau\tau_c = \frac{xc\tau_c\lambda^2}{4\pi n_0^2\Delta V} \left( \frac{\ln 2}{\pi} \right)^{1/2}$$

and further

$$\zeta = \frac{\tau}{2\alpha}$$

Substituting these quantities into these equations leads to a quadratic in  $W_p$

$$W_p^2(1-\zeta)\tau_B - W_p\zeta - \frac{1+\zeta}{\tau} = 0 \quad (10)$$

This has only one physically acceptable solution:

$$f(\text{threshold}, QDXX) = \frac{W_p}{\sigma_A}$$

$$= \frac{\tau + \sqrt{\tau^2 + \frac{4(4\alpha^2 - \tau^2)\tau_B}{\tau}}}{2\sigma_A(2\alpha - \tau)\tau_B} \quad (11)$$

We therefore find that the spontaneous emission lifetime  $\tau$  does indeed play a very important role in deciding the threshold fluence of a quantum dot laser. It is worthwhile to compare this result to the behavior of ideal three and four

level systems (Figure 1 c, d). These familiar results may also be cast into a similar, lifetime dependent form (details are available in the supplementary section):

$$f(\text{threshold}, 3\text{-level}) = \frac{\alpha + \tau}{\sigma_A(\alpha - \tau)\tau} \quad (12)$$

and

$$f(\text{threshold}, 4\text{-level}) = \frac{\tau}{\sigma_A(\alpha(\tau - \tau_D) - \tau^2)} \quad (13)$$

Here  $\tau_D$  is the lifetime of the  $n_1$  level.

In each case, we observe that at a sufficiently high spontaneous lifetime, the threshold fluence needed for lasing becomes infinite. For example, in the case of a three level system, this corresponds to the situation when  $\tau = \alpha$ . Equations (11)-(13) are thus very similar in terms of their behavior towards the longer lifetime side. It is however important to consider one other potential situation involving QDs, where the behavior is strikingly different.

In deriving equation (11), we have considered that biexcitons cause gain while single excitons are optically transparent. This need not always be true. For example, in certain QDs, single excitons have been reported to give rise to gain<sup>14,15</sup>. This could occur either because of a four-level like energy level scheme where the ground state is optically transparent or else due to the inability of single excitons to absorb band edge light.

Regardless of the microscopic origins of such a situation, it implies that a QD with a single exciton needs to be considered as inverted, while a biexciton becomes optically inconsequential. Transparency of the medium thus occurs when the gain from single excitons balances the loss from unoccupied QDs, i.e.  $N = 2(1-N-B)$ . We notice that even though biexcitons do not cause gain, they still help define the threshold criterion. This occurs because the presence of a biexciton in the system also implies the availability of a reduced number of single excitons. The equation for band edge optical flux now becomes:

$$\frac{d\phi}{dt} = c\sigma\phi x N - 2c\sigma\phi x(1-N-B) - \frac{\phi}{\tau_C} \quad (14)$$

The same treatment as before leads to

$$f(\text{threshold}, QDX) = \frac{(\alpha - \tau) \pm \sqrt{(\alpha - \tau)^2 - 4(\tau + 2\alpha)\tau_B}}{2\sigma_A\tau\tau_B} \quad (15)$$

This equation is rather different from equations (11)-(13), particularly in the sense that there is no singularity at higher spontaneous lifetimes. It is however important to interpret this result with caution. We firstly observe that both roots of

this equation are physically allowed. Therefore, it is possible for this system to have two thresholds. Using equation (8), it is simple to see that the lower threshold corresponds to a low population of biexcitons, while the upper threshold corresponds to a higher population of biexcitons. This feature becomes more apparent when we consider actual material parameters. We also note that these equations consider a unit quantum yield. The impact of sub-unity quantum yields is discussed in the supplementary section.

## Results and Discussion

For solid-state laser materials, we employ a refractive index of 1.82 and a doping level of  $10^{20}/\text{cm}^3$  for each crystal. For QDs, a medium refractive index of 1.3 (toluene) and a number density of  $10^{18}/\text{cm}^3$  is assumed. The bandwidth,  $\Delta\nu$  is taken to be  $1.2 \times 10^{11}$  Hz for Nd:YAG,  $10^{14}$  Hz for Ti:Sapphire,  $3.3 \times 10^{11}$  Hz for Ruby and  $10^{13}$  Hz for QDs. For ease of comparison, we assume that each material has a band center at  $10^3$  nm. Finally, we take  $\tau_B = 700$  ps and  $\tau_D = 10$  ps. The absorption cross sections of all solid state materials are taken to be  $10^{-19}$  cm<sup>2</sup>. For QDs, the cross section is taken to be  $2 \times 10^{-14}$  cm<sup>2</sup>. Pump Photon energies are 1.5 eV for Nd:YAG, 2.3 eV for Ti:Sapphire and Ruby and 3 eV for QDs.

The value of  $\alpha$  is also dependent on the characteristic time  $\tau_C$  of photons inside the cavity. While amplified spontaneous emission (ASE) does not involve an actual resonator, it does correspond to photons spending a finite amount of time within the gain medium. We consider the situation where photons on average travel through a nominal path length of 1 mm in the gain medium. The effect of a resonator is to increase the characteristic time of photons in the medium. Here we consider that a resonator provides a 100 fold enhancement. With these parameters, we obtain that in the case of ASE  $\alpha = 2.2 \mu\text{s}$  for QDs,  $\alpha = 9000 \mu\text{s}$  for Nd:YAG,  $\alpha = 9 \mu\text{s}$  for Ti:Sapphire and  $\alpha = 3000 \mu\text{s}$  for Ruby (Cr: Sapphire).

Figure 2a plots the threshold fluence for single exciton gain as a function of the threshold lifetime. Plotted in this figure are the two different physical solutions (+ and -) of equation (15), versus the spontaneous lifetime. It is clear that at longer lifetimes both the solutions approach a common point. Subsequently, the expression under the square root becomes imaginary and no lasing is possible.

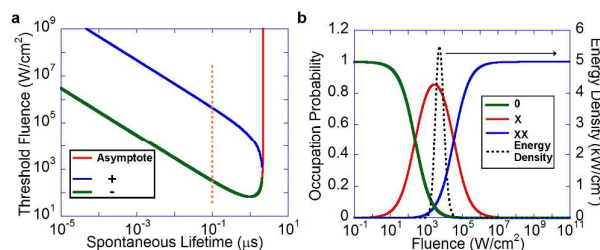
Figure 2a also shows a dashed line at a spontaneous lifetime of  $0.1 \mu\text{s}$ . The variation of excitonic populations as a function of fluence along this curve is plotted in Figure 2b. The physical meaning of two thresholds becomes clear from Figure 2b. At low fluences, most QDs are in their ground states, while very few dots have excitons. As the fluence is increased, the ground state population falls, while the population of QDs in the first excited state increases. At still higher fluences, the populations in the ground state fall further, while the number of biexcitons in the system increases. It is important to note that beyond a

certain fluence, the number of single excitons in the system begins to fall with increasing fluence. Since the extent of inversion in this case depends only on the number of single excitons, this quantity falls at very large fluences. This has a very unusual implication in the case of lasing under the single exciton regime, where beyond a certain threshold, laser output once again ceases (given by the photon energy density (dashed line, figure 2b)). The existence of two thresholds thus has a clear interpretation. While the lower threshold corresponds to the onset of lasing, the upper threshold occurs when the pump fluence is high enough to pump a very substantial number of biexcitons. Under these circumstances, there is an insufficient number of single excitons present to overcome ground state absorption, and so lasing stops. The second threshold therefore represents an upper bound to the lasing fluence. At a sufficiently high spontaneous lifetime, the expression under the radical sign first vanishes and then becomes imaginary.

This corresponds to a situation where the spontaneous lifetime is so large that the biexcitonic population becomes substantial even before the onset of lasing. Thus, no real threshold exists for sufficiently long spontaneous lifetimes. It is further instructive to consider the  $\tau_B \rightarrow 0$  asymptote for equation (15). This is given by is given by

$$f_0(\text{threshold}, QDX) = \frac{(2\alpha + \tau)}{\sigma_A \tau (\alpha - \tau)} \quad (16)$$

This is similar to the threshold of a perfect three level system (Equation (12)). The factor of 2 arises because in a QD system, two singly excited QDs are needed to overcome losses of a single QD in the ground state. This equation is also plotted in Figure 2a. Numerically, this equation is essentially identical to the lower threshold. At longer lifetimes, while equation (15) becomes imaginary, its asymptotic form exhibits a singularity. The consideration of the zero biexcitonic lifetime asymptote thus does not have any significant bearing on the lower lasing thresholds of the QD materials.

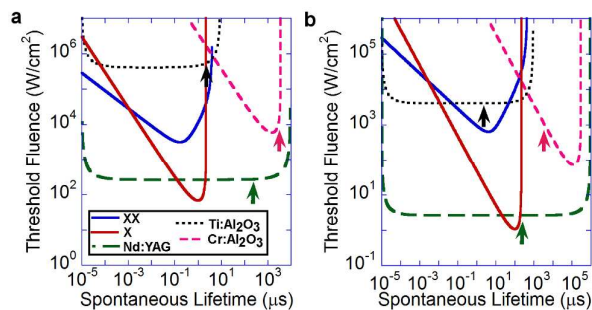


**Figure 2.** a. Variation of threshold fluence for QD ASE under the single excitonic gain regime. The asymptote is given by equation 16. b. Variation of occupation probability with fluence for QD ASE. QDs have a lifetime of  $0.1 \mu\text{s}$ , indicated by the dashed line in panel a. Also shown is the energy density of photons as a function of fluence (dashed).

The above kinetic models for quantum dots with single exciton and biexciton gain suggests that the fluence required for attaining optical transparency is a strong function of spontaneous lifetime, other parameters being constant. This



enforces our claim that stretching excitonic spontaneous lifetimes by several orders of magnitude could help attain gain or laser action at very low fluences.



**Figure 3.** The relationship between threshold fluence and spontaneous lifetime for various laser materials for **a.** ASE and **b.** a resonator. The actual spontaneous lifetimes of Ti: Sapphire, Ruby and Nd:YAG are invariant and are indicated by an arrow. The lifetimes of QDs are tunable and are indicated by solid lines. X corresponds to equation 16, while XX corresponds to equation 11.

Figure 3a plots the thresholds of various systems as a function of spontaneous lifetime (Equations (11)–(13) and (16)). Several important trends are immediately apparent. For sufficiently long spontaneous lifetimes, none of the systems are able to achieve threshold at any pump fluence. This occurs because the stimulated emission cross section of the material becomes too small for the material to overcome cavity losses regardless of extent of population inversion. While four level systems are rather insensitive to spontaneous lifetimes, three level materials such as ruby, as well as quantum dots are highly sensitive to this parameter.

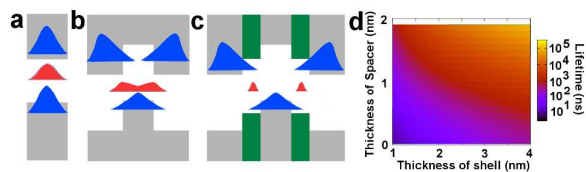
In particular both QDs as well as Ruby show a strong minimum in the gain threshold at particular values of spontaneous lifetimes. In the case of QDs operating under biexcitonic gain, this minimum occurs at lifetimes as long as 600 ns for ASE. QDs with single exciton gain have a much lower gain threshold that materializes at lifetimes around 2  $\mu$ s. In contrast, the optimal spontaneous lifetime of a ruby-like material is seen to be 1 ms for purposes of ASE. It is also important to comment that at their optimal lifetimes, QDs can potentially outperform even established laser materials such as Nd:YAG; this remarkable observation is simply a reflection of their significantly higher optical cross sections. The introduction of a resonator reduces thresholds significantly, however the qualitative aspects remain unchanged. Equations (11)–(13) and (15) are fairly general and are applicable to other systems as well. For example, ASE as well as lasing have been recently observed in colloiddally grown 2-dimensional quantum wells<sup>16</sup>. Using parameters reported in reference<sup>16</sup> ( $\Delta\nu = 1 \times 10^{13}$  Hz,  $\tau_B = 124$  ps,  $\tau = 5.1$  ns, refractive index 1.7, cross section of  $5 \times 10^{-14}$  and a packing density of  $1.5 \times 10^{18}$  cm<sup>-3</sup>) we obtain  $\alpha = .5 \mu$ s which implies a threshold of 60 W/cm<sup>2</sup> for ASE. This is in excellent agreement with the experimentally observed threshold values of 10–400 W/cm<sup>2</sup>.

At this point it is also important to realize that the curves not shown by solid lines in figure 3 are essentially hypothetical; it is impossible to tune the spontaneous lifetime of a solid state laser material to the extent shown. The arrows shown in the figure represent the actual spontaneous lifetimes of each of these materials. Because of the lack of dependence of a four level system threshold on spontaneous lifetimes, Nd:YAG as well as Ti:Sapphire remain near optimal regardless of the oscillator. The ruby crystal however has a less than optimal performance for the oscillator shown in figure 3b. In contrast, to other solid state laser materials exemplified here, it is indeed possible to tune the spontaneous emission lifetimes of QDs even to the extent required by this application. This is an advantage that QDs offer even against 2 dimensional semiconductors.

Several routes to achieve long lifetimes<sup>17</sup> in QDs already exist however, none of these are conducive for lasing. A IV-VI compound such as PbSe<sup>18</sup> exhibits spontaneous lifetimes that are appropriate for low threshold gain, however the high band edge degeneracy<sup>19</sup> ensures that the onset of gain would only occur at  $N \geq 4$ . Multi carrier recombination<sup>20</sup> has thus prevented the observation of lasing under continuous wave excitation in this system. Alternatively, II-VI QDs also exhibit microsecond long lifetimes at low temperatures<sup>21, 22</sup>. The effects of temperature on QDs are simple to explain, and as it turns out, are not useful from the gain application viewpoint. QD emission in II-VI QDs slows down at low temperatures due to the depopulation of the emissive bright state. Thus, although there is indeed a slowing of the emission, the lifetimes of individual emissive states are entirely unchanged and also non optimal for lasing. An additional complication at low temperatures is the narrowing of the homogenous linewidth<sup>10, 23, 24</sup> that reduces the number of QDs capable of emission at certain wavelengths.

In light of the above examples, a more complete list of requirements for low threshold gain may be drawn up. Low threshold gain requires an ensemble composed of QDs with reasonable quantum yields, a two-fold degenerate band edge, microsecond long spontaneous emission lifetimes, as well as comparable homogenous and inhomogenous linewidths. While no QDs that satisfy these criteria exist till date, it is indeed possible to show that a suitably designed heterostructure could fulfil these requirements.

In an effective mass description, a long spontaneous lifetime can arise from poor overlap between electron and hole envelop functions. Traditional single component as well as type-I and type-II QDs<sup>25</sup> exhibit electron-hole overlaps (Figure 4 a, b) that are high enough to inhibit lifetime regulation in any reasonably sized QD<sup>5</sup>. We show that the introduction of an intermediate layer of semiconductor allows for significant reduction in electron-hole overlap and the consequent tuning of the emission lifetimes of staggered offset materials into the microsecond regime (Figure 4c). Calculations of lifetime that



**Figure 4.** Electron hole overlap in a. single component QDs, b. type-II QDs and c. QDs with a spacer. d. Variation of QD lifetimes with spacer and shell thickness under a k.p approximation.

we show here rely on a Bastard model<sup>26</sup>. The valence band was assumed to have a parabolic dispersion, while the non-parabolicity of the conduction band is accounted for in a parametric manner. The allowed envelop functions were determined by neglecting the interactions of the conduction band functions to other remote functions and then ensuring the continuity of the envelope  $\varphi(\vec{r}, \varepsilon)$  and the associated current  $\varphi'/m(\varepsilon, \vec{r})$  across each interface. The minimum values of  $\varepsilon$  satisfying these criteria give rise to the allowed band edge envelop functions. The envelope functions calculated within this approximation enable one to estimate the overlap integrals of the conduction and valence band edge. The lifetimes of the excitons are proportional to the inverse square of the overlap. The absolute values of lifetimes were obtained by fixing a lifetime of 10 ns in absence of the intermediate spacer layer. Other lifetimes were then obtained from the ratio of the squares of overlap integrals between conduction and valence states. Band offsets as well as effective masses and band gaps for various layers were taken from literature. The ZnTe/ZnSe/CdS conduction band offsets<sup>27, 28</sup> were taken to be +1.2/+0.9/0. The electronic effective masses of these materials are taken to be 0.21/0.21/0.2, while the hole effective masses are taken to be 0.2/0.6/0.8<sup>5, 27</sup>.

We find that the introduction of a ZnSe spacer between the ZnTe and CdS regions leads to the predictable lengthening of the lifetimes of this material. The magnitude of this effect is rather impressive. Figure 4d presents a two dimensional plot of the predicted excitonic lifetime as a function of shell and spacer thickness. For a simple type-II structure (zero spacer thickness), a lifetime higher than 10 ns appears unachievable. However, even for a spacer as thick as 1 nm, suitable shell thicknesses could allow for the tuning of spontaneous lifetimes to values as long as 1  $\mu$ s. This is more than sufficient to allow QDs to exhibit lasing behavior at extremely low fluences, well below 100 W/cm<sup>2</sup>.

## Conclusion

QD lasing under continuous wave pumping has been a long elusive goal. While much attention has been devoted to the development of QDs with controlled biexcitonic decay, we show that regulated spontaneous decay is of even greater importance. We show that control over spontaneous decay could be straight forwardly achieved by the introduction of a

wide band gap spacer layer within a type-II structure. Based on kinetic modeling of a QD laser, this simple transformation is expected to allow QDs to lase at extremely low fluences, corresponding to 100 W/cm<sup>2</sup> or lower. Attainment of laser action from a QD material under continuous wave pumping is of special significance. Unlike conventional solid state laser materials, QDs exhibit extremely broad absorption bands that cover all visible wavelengths higher than the band gap. This broadband absorption could potentially allow such QDs to convert light from diffuse sources such as sunlight into a laser beam. While this possibility has been demonstrated in solid state materials such as Nd:YAG<sup>29</sup>, such demonstrations are still very challenging experimentally and represent the state of the art in solar concentration as well as materials design. When using spectrally broad pump sources such as sunlight, QD lasers compare favorably to existing materials because of their large excitation cross sections as well as their broad pump bands. The development of QDs that lase under continuous wave excitation will open up novel applications for lasers in light concentration and transmission that are unfeasible in the current scenario.

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**Novelty:** We discuss approaches that could lead to very low threshold continuous wave quantum dot lasers.

TOC Graphic:

