## **Excitonic exchange effects on the radiative decay time of monoexcitons and biexcitons in quantum dots**

Gustavo A. Na[r](#page-5-1)va $\alpha$ ez,  $\ast$  Gustavo Bester, Alberto Franceschetti, and Alberto  $\alpha$  $N_{\epsilon, \epsilon}$  and  $R_{\epsilon}$  in  $E$  in  $L_{\epsilon}$  and  $\epsilon$  in  $G$  is a colorado 80401, USA  $(\sum_{1} 30 + 2006; 1)$  17 November 2006)  $\mathbf{E}_{\text{inert}}$ estron-hole exchange interaction splits the exciton ground state into  $\mathbf{E}_{\text{inert}}$  states. The dynamics of those states dependence on the internal relaxation time between bright and dark states spin-flip t and on the radiative recombination time of the bright states. On the other hand, the calculated values of these recombination times depend not only on the treatment of correlation effects, but also on the accuracy of the electron and hole wavefunctions. We calculate the radiative decay rates for monoexcitons and biexcitons in  $\Gamma$  , As )  $G$  /Ga  $A$  self-assembled and colloidal  $C$  and colloidal wave functions. The correlations for  $G$ We show how the radiative decay time *<sup>R</sup>* <sup>0</sup>- of the monoexciton depends on the spin-flip relaxation time between bright and dark states. In contrast, a biesciton has no bright-dark splitting, so the decay time of th biexciton *<sup>R</sup>XX*0- is insensitive to this spin-flip time. This results in ratios *<sup>R</sup>* <sup>0</sup>-/ *<sup>R</sup>XX*0- of 4 in the case of for an ratio of 2 in the case of slow spin flip. For  $\H$  ,  $G$  )A /Ga s, we compare our results with the model calculation of  $\mathcal{M}$   $\left[ \ldots \right]$  **B 73**, 165305 (2006). When the same spin-flip rates are spin-flip rates assumed, our predicted *<sup>R</sup>* <sup>0</sup>-/ *<sup>R</sup>XX*0- agrees with that of Wimmer *et al.*, suggesting that our treatment of correlations is adequate to predict the ratio of monoexciton and biexciton radiative lifetimes. Our results agree well with experiment on self-assembled dots when assuming slow spin flip.  $\mathbf{C}$  is assuming spin flip for colloidal  $\mathbf{c}$ dots the agreement with experiment is best for fast spin flip. D I:  $10.1103 / P$  B.74.205422  $AC_{-1}$  ():  $73.21.\frac{1}{2}$ ,  $71.35.$ ,  $78.60.$ **I. INTRODUCTION: RELATION BETWEEN APPARENT AND MICROSCOPIC CARRIER DECAY** We address here the subject of how to compare measured  $\omega$ exciton  $R$  (  $^{(0)}$  ) and biexciton  $R$  (  $^{(0)}$  )  $_{\sim}$  and  $_{\sim}$   $_{\sim$ times with calculated values.  $\mathbf{E}_{\rm acc}$  is an ensemble of of ensemble of offs  $\mathbf{E}_{\rm acc}$ quantum dots is excited by an optical pump-pulse and the  $p_{1665} = 1665 \text{ D} 3011665 -3011665 -3011665-\frac{3}{14}3011665-\frac{1}{14}5011665-\frac{1}{14}5911665-\frac{1}{14}59105-\frac{1}{14}5910459.1$  $[F_1, 1]$ .  $1/3$  $1/3$  **F**<sub>c</sub>  $\ldots$   $C_{2v}$  symmetry of  $C_{2v}$  $\left( \mathrm{I} \ \, , \mathrm{G} \ \, \right)$ As/ $\mathrm{G} \ \, \mathrm{A}$ self-assembled quantum dots, the four states are the high-energy bright state -*d* splitting between the dark and the bright states is an order of magnitude larger than in self-order of  $\tau$ assembled dots, ranging from 2–20 meV. [4](#page-5-4)[,5](#page-5-5) I<sub>n both</sub> self-2 2<br>0 0 منعت فيال لا يكون كون بينومبية <sub>إ</sub>كتا<sub>ل</sub>ين في كيار state has no fine structure and corresponds to a single bright state that can decay to the four states of <sup>0</sup> 1 0 <sup>1</sup> in the monoex- $[F_{\cdot}, 1(\cdot)].$  $[F_{\cdot}, 1(\cdot)].$  $[F_{\cdot}, 1(\cdot)].$ In this paper we show that is denoted and fine-due to exchange and  $\epsilon$  . Figure [1](#page-1-0) $\Gamma$  , and bien energy the monocycle  $\epsilon$ structure in the monoexciton, the measured apparent radia-levels that enter our calculations. We do not consider highertive recombination  $\mathbb{R}^{\left( { - 0} \right)}$  depends on the bright-to-dark  $s \sim \tau$  is the  $s$  -  $B$ D with  $R_{BD} = \frac{1}{BD}$ ,  $B \to \infty$   $t_{\perp}$  . atomistic pseudopotential-based approach combined with the configuration-interaction method[,6](#page-5-6) we calculate the characteristic radiative recombination rates between *B* and the ground state *RB*0- and between *D* and the ground state *RD*0 and input them in a set of rate equations with varying *RBD* rates. We find that the photon emission rate decays as a single-exponential with rate  $R_{B0}$  for spin flip times,  $\frac{1}{2}$  for slow spin flip times,  $\frac{1}{2}$ as a biexponential for intermediate *BD*, and as a single- $R_{B0}/2$  for fast spin flip times.  $R_{B0}/2$  for fast spin flip times.  $\binom{n}{k}$ سم <sub>ال</sub> المعلوم بالمعادل المعادل المعادل المعادل المعرض المعادل المعادل المعادل المعادل المعادل المعادل المعادل <br>المعادل المعادل الم culate the characteristic recombination rates  $R_{0B}$  and  $R_{0D}$  of  $R_{0D}$  of  $\overline{R}_{0D}$ the biestim ground state into the bright and dark states of  ${\bf k}$  and data states of the monoton monoton. We find that  $R_{0B} \simeq R_{B0}$  and  $R_{0D} \simeq R_{D0}$ that the biexciton radiative decay is a single exponential with  $t$  is a single exponential with  $\alpha$  $\therefore$  2*R*<sub>0</sub><sup>B</sup> −1</sub>  $\therefore$   $\binom{R_{BD}}{k}$ . iii) that due to the aforementioned dependence of the monoexcito decay time  $R_{BD}$ , the ratio  $R \begin{pmatrix} 0 \end{pmatrix} I_R \begin{pmatrix} 0 \end{pmatrix} = 0$  $v_{\rm eff}$  (  $v_{\rm eff}$  and  $2$  for the limiting cases of fast and slow spinners flip, respectively. We thus resolve the apparent contradiction between the recent model calculations of Wimmer and  $c_{1}$  **c**  $\mathbf{k}$   $\mathbf{k}$ ,  $\mathbf{k}$ <sup>7</sup> who found  $\mathbf{k}$   $\mathbf{k}$   $\mathbf{k}$   $\mathbf{0}$   $\mathbf{k}$   $\mathbf{k}$   $\mathbf{0}$   $\mathbf{k}$   $\mathbf{0}$   $\mathbf{k}$   $\mathbf{k}$  ous atomistic  $\gamma$  and  $\gamma$  and  $\gamma$  and  $\gamma$  and  $\gamma$   $\gamma$   $\gamma$   $\gamma$   $\gamma$   $\gamma$   $\gamma$  $R$ <sup>2</sup> (0)/ $R$ <sup>3</sup> (0)/ $R$ <sup>2</sup> (0) = 4. illustrate our findings with atomistic, pseudopotentialbased calculations for a prototypical self-assembled I<sub>0.6</sub>G<sub>0.4</sub>As/Ga<sub>s</sub> dot and a C<sub>dSe</sub> colloidation and a co  $\label{eq:4} \mathcal{L}(\omega) = -\left|\frac{1}{\lambda}\right|^2\left|\frac{1}{\lambda}\right|^2\left|\frac{1}{\lambda}\right|^2.$ **II. RATE EQUATIONS FOR THE RADIATIVE DECAY OF THE MONOEXCITON**

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<span id="page-1-0"></span>
$$
L_{\bullet} = (R_{\bullet} + R_{\bullet} + R_{\bullet} + R_{\bullet} + R_{\bullet}) + R_{\bullet} + R_{
$$

*dndt*

lying states because most time-resolved photoluminescence experiments are conducted at temperatures such that the occupation of the those states is negligible. From  $\mathrm{F}_J$  ,  $\mathrm{1}\mathrm{I}_\mathrm{c}$  $\mathrm{1}\mathrm{I}_\mathrm{c}$  $\mathrm{1}\mathrm{I}_\mathrm{c}$  ) and see that there are a number of discrete transitions channels denoted below by rates *Rij*. We next set up a set of channelspecific rate equations describing how the individual levels of  $F_2$  ,  $\mathbf{1}$  $\mathbf{1}$  $\mathbf{1}$   $\left(\begin{array}{ccc} 0 & 0 & 0 \\ 0 & 0 & 0 \end{array}\right)$  from which we will deduce the will de global decay of the ground state <sup>0</sup>*t*- which is measured.  $U$ sing the characteristic radiative radiative radiative radiative rates of the four excitonic states of <sup>0</sup> 1 0 1 and the ground state <sup>0</sup> 0 0 0 we establish the  $f_{\rm eff}$  system of  $f_{\rm eff}$  system of  $f_{\rm eff}$  system of  $f_{\rm eff}$ 

<span id="page-1-1"></span>
$$
L_{\nu} / \sqrt{1 + R_{\nu}} = (R_{\nu} / + R_{\nu} + R_{\nu} / + R_{\nu}) + R_{\nu} / + R_{\nu}
$$
  
+  $R_{\nu} / \sqrt{1 + R_{\nu}}$ 

$$
L_{\nu} = (R_{\nu} + R_{\nu} + R_{\nu} + R_{\nu} + R_{\nu}) + R_{\nu} + R_{\nu} + R_{\nu}.
$$
  
+ R\_{\nu} ,

## <span id="page-2-0"></span>then solve  $\Xi$  and  $\overline{\Theta}_+$  and  $\overline{\Xi}_+$  and the photon emission rate the photon  $\Xi$  $I(t) = R_{B0}^T - B(t) + R_{D0} - D(t)$ , which is directly comparable to  $A = -1$ time-resolved photoluminescence PL- experiments.

## **III. RATE EQUATION FOR THE RADIATIVE DECAY OF THE BIEXCITON**

The biexciton has a nondegenerate state without *B*-*D*

<span id="page-3-0"></span>
$$
F = \frac{1}{2} \left( \frac{R_{B0}}{F} - \frac{s}{s} \right),\tag{12}
$$

$$
S = \frac{1}{2} \left( \frac{R_{B0} - F}{F - s} \right). \tag{13}
$$

In time-resolved PL experiments the measured signal *It* is proportional to the number of photons per unit of time: *dn*<sup>0</sup> /*dt* Eq. [6](#page-1-1)-, which under the assumption of *RD*0=0 results in *It*-=*RB*<sup>0</sup> *<sup>B</sup>t*-. Note that by being proportional to the occupation of the bright state the signal *It* carries information on [b](#page-3-0)oth radiative and nonradiative spin flipprocesses.

