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

Gustavo A. Narvaez,* Gabriel Bester, Alberto Franceschetti, and Alex Zunger†
National Renewable Energy Laboratory, Golden, Colorado 80401, USA
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Electron-hole exchange interaction splits the exciton ground state into “dark” and “bright” states. The dynamics of those states depends on the internal relaxation time between bright and dark states (spin-flip time), 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 (In,As)Ga/GaAs self-assembled and colloidal CdSe quantum dots from atomistic correlated wave functions. We show how the radiative decay time \( \tau_R(X^0) \) of the monoexciton depends on the spin-flip relaxation time between bright and dark states. In contrast, a biexciton has no bright-dark splitting, so the decay time of the biexciton \( \tau_R(XX^0) \) is insensitive to this spin-flip time. This results in ratios \( \tau_R(X^0)/\tau_R(XX^0) \) of 4 in the case of fast spin flip, and a ratio of 2 in the case of slow spin flip. For (In,Ga)As/GaAs, we compare our results with the model calculation of Wimmer et al. [Phys. Rev. B 73, 165305 (2006)]. When the same spin-flip rates are assumed, our predicted \( \tau_R(X^0)/\tau_R(XX^0) \) 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 quantum dots when assuming slow spin flip. Conversely, for colloidal dots the agreement with experiment is best for fast spin flip.

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I. INTRODUCTION: RELATION BETWEEN APPARENT AND MICROSCOPIC CARRIER DECAY

We address here the subject of how to compare measured exciton \( \tau_R(X^0) \) and biexciton \( \tau_R(XX^0) \) radiative relaxation times with calculated values. Experimentally, an ensemble of quantum dots is excited by an optical pump-pulse and the photons subsequently emitted are counted as a function of time. The photon emission rate vs time is often not a single exponential. The reason for this is that even in a single dot the monoexciton ground state is not a single state but a manifold of exchange and fine-structure split states with internal carrier dynamics. In III-V and II-VI dots the monoexciton ground state originates from the \( C_{2v} \) symmetry of (In,Ga)As/GaAs self-assembled quantum dots, the four states are the high-energy bright state \( B \) which consists of a pair \( b \) and \( b' \) split by a few \( \mu eV \) and the low-energy dark state \( D \) which consists of a pair \( d \) and \( d' \) that is quasidegenerate. \( B \) and \( D \) are split by a few \( \mu eV \) due to exchange effects. For colloidal CdSe quantum dots with the \( C_{6v} \) symmetry the internal \( d-d' \) and \( b-b' \) splittings of the two pairs are small, with the \( b-b' \) recently measured to be about 1–2 meV.\(^4\) In turn, the \( b-d \) splitting between the dark and the bright states is an order of magnitude larger than in self-assembled dots, ranging from 2–20 meV.\(^5\) In both self-assembled and colloidal dots, the biexciton ground state \( e_0^2|h_0^0 \) state has no fine structure and corresponds to a single bright state that can decay to the four states of \( e_0^1|h_0^0 \) in the monoexciton [Fig. 1(c)].

In this paper we show that (i) due to exchange and fine-structure in the monoexciton, the measured apparent radiative recombination time \( \tau_R(X^0) \) depends on the bright-to-dark spin-flip relaxation time \( \tau_{BD} \) with rate \( R_{BD} = \tau_{BD}^{-1} \). By using an atomistic pseudopotential-based approach combined with the configuration-interaction method,\(^5\) we calculate the characteristic radiative recombination rates between \( B \) and the ground state \( (R_{B0}) \) and between \( D \) and the ground state \( (R_{D0}) \) and input them in a set of rate equations with varying \( R_{BD} \) rates. We find that the photon emission rate decays as a single-exponential with rate \( R_{B0} \) for slow spin flip times, as a biexponential for intermediate \( \tau_{BD} \), and as a single-exponential with rate \( R_{B0}/2 \) for fast spin flip times. (ii) Within the same approach used for the monoexciton, we calculate the characteristic recombination rates \( R_{D0} \) and \( R_{BD} \) of the biexciton ground state into the bright and dark states of the monoexciton. We find that \( R_{D0} = R_{B0} \) and \( R_{D0} \approx R_{DD} \) and that the biexciton radiative decay is a single exponential with a decay time \( \tau(XX^0) \approx 2R_{B0}^{-1} \) regardless of \( R_{BD} \). (iii) We show that due to the aforementioned dependence of the monoexciton decay time on \( R_{BD} \), the ratio \( \tau_R(X^0)/\tau_R(XX^0) \) has the values of 4 and 2 for the limiting cases of fast and slow spin flip, respectively. We thus resolve the apparent contradiction between the recent model calculations of Wimmer and co-workers,\(^7\) who found \( \tau_R(X^0)/\tau_R(XX^0) = 2 \), and our previous atomistic-based realistic calculations of \( \tau_R(X^0) \) and \( \tau_R(XX^0) \) in which we found\(^8\) \( \tau_R(X^0)/\tau_R(XX^0) = 4 \).

We illustrate our findings with atomistic, pseudopotential-based calculations for a prototypical self-assembled In,As/Ga,GaAs dot and a CdSe colloidal dot, comparing with available data.

II. RATE EQUATIONS FOR THE RADIATIVE DECAY OF THE MONOEXCITON

Figure 1(a) shows the monoexciton and biexciton energy levels that enter our calculations. We do not consider higher-
Using the characteristic radiative rates of the four excitonic ground state \( e_0^0 \) and \( e_0^0 \), Thin- and thick-line ellipses indicate, respectively, nonradiative thermalization as well as spin-flip channels.

The model three-level system for the bright-dark splitting is the \( R_{bd}^{RDB} = R_{bd}^{RBD} \) rate of the biexciton in GaAs dots allows us to make the assumption \( R_{bb}^{RBB} = R_{bb}^{RBD} \). For CdSe colloidal dots the bright-dark splitting is about an order of magnitude larger and therefore \( R_{bb}^{RBD} \) for \( T \geq 300 \) K. (In the results discussed in Sec. V we adopt the regime in which \( R_{bb}^{RBB} \).)

Assumption (i) is justified in different range of temperatures determined by the magnitude of the small splittings between \( b-b' \) and \( d-d' \), \( T \geq 2 \) K in self-assembled dots and \( T \geq 20 \) K in CdSe colloidal dots. Regarding (ii), the small fine-structure bright-dark splittings \( (10-300 \mu eV) \) in (In,Ga)As/GaAs dots allows us to make the assumption \( R_{bb}^{RBB} = R_{bb}^{RBD} \). For CdSe colloidal dots the bright-dark splitting is about an order of magnitude larger and therefore \( R_{bb}^{RBD} \) for \( T \geq 300 \) K. (In the results discussed in Sec. V we adopt the regime in which \( R_{bb}^{RBB} \).)

Assumption (ii) is assumed to be independent on the index of the bright or dark state while keeping the distinction between bright-dark and dark-bright transition rates

\[
R_{bb}^{RBB} = R_{bb}^{RDD} = R_{bb}^{RBD},
\]

and (iii) the decay rate of \( b \) and \( b' \) to \( e_0^0 \) is equal, and so are the decays of \( d \) and \( d' \) to \( 0 \)

\[
R_{dd}^{RBD} = R_{dd}^{RDD}, \quad R_{dd}^{RDD} = R_{dd}^{RDD}.
\]

The five-level system of rate equations [Eq. (1)] that describe the radiative decay of \( X^0 \) can be reduced to a three-level system when (i) the thermalization rate [Fig. 1(a)] within the \( b-b' \) bright and within the \( d-d' \) dark states is assumed equal

\[
R_{bh}^{RBD} = R_{bh}^{RDD} = R_{bh}^{RDD},
\]

(ii) spin-flip rates between the bright and the dark states [Fig. 1(a)] are assumed to be independent on the index of the bright or dark state while keeping the distinction between bright-dark and dark-bright transition rates

\[
R_{bd}^{RBD} = R_{bd}^{RDD} = R_{bd}^{RDD},
\]

\[
R_{db}^{RBD} = R_{db}^{RDD} = R_{db}^{RDD},
\]

and (iii) the decay rate of \( b \) and \( b' \) to \( e_0^0 \) is equal, and so are the decays of \( d \) and \( d' \) to \( 0 \)

\[
R_{dd}^{RBD} = R_{dd}^{RDD} = R_{dd}^{RDD}.
\]

We next set up a system of channel-ellipses indicate, respectively, nonradiative thermalization as well as spin-flip channels. (b) The model three-level system for the monoexciton decay. \( R_{bd}^{RBD} \) is the bright-dark (dark-bright) rate while \( R_{bd}^{RBD} \) and \( R_{dd}^{RBD} \) are radiative decay rates for the bright and dark (model) state, respectively. (c) The same as (a) for the biexciton ground state \( e_0^0 \) and \( e_0^0 \), and (d) model representation, with radiative decay rates \( R_{bb}^{RBB} \) and \( R_{dd}^{RDD} \).
TABLE I. Calculated values for the radiative characteristic rates $R_{i0}$ and $R_{i} (i=b, b', d, d')$ for the monoexciton ($X^0$) and biexciton ($XX^0$), respectively, in an alloyed In$_{0.4}$Ga$_{0.6}$As/GaAs dot (base diameter $b$ =252 Å and height $h$= 35 Å) and a CdSe colloidal dot (diameter $D$=38 Å). The approximate rates $R_{b0}$, $R_{d0}$ that enter the model three-level system of rate equations [Eq. (6)] are also shown, as well as the rates $R_{b0}$ and $R_{d0}$ for the biexciton.

<table>
<thead>
<tr>
<th></th>
<th>In$<em>{0.4}$Ga$</em>{0.6}$As/GaAs</th>
<th>CdSe</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$X^0$</td>
<td></td>
</tr>
<tr>
<td>$R_{b0}$</td>
<td>0.89 ns$^{-1}$</td>
<td></td>
</tr>
<tr>
<td>$R_{b'/0}$</td>
<td>0.91 ns$^{-1}$</td>
<td></td>
</tr>
<tr>
<td>$R_{d0}$</td>
<td>$6.65 \times 10^{-5}$ ns$^{-1}$</td>
<td>$R_{d0}=0$</td>
</tr>
<tr>
<td>$R_{d'/0}$</td>
<td>$6.3 \times 10^{-6}$ ns$^{-1}$</td>
<td>$R_{d'/0}=0$</td>
</tr>
<tr>
<td>$XX^0$</td>
<td>$0.83 \times 10^{-6}$ ns$^{-1}$</td>
<td>$R_{bb}=0.84$ ns$^{-1}$</td>
</tr>
<tr>
<td>$R_{bb}$</td>
<td>$0.85$ ns$^{-1}$</td>
<td>$R_{bb'}=0.13$ ns$^{-1}$</td>
</tr>
<tr>
<td>$R_{bb'}$</td>
<td>$0.67 \times 10^{-3}$ ns$^{-1}$</td>
<td>$R_{bb'}=0$</td>
</tr>
<tr>
<td>$R_{bd}$</td>
<td>$0.46 \times 10^{-4}$ ns$^{-1}$</td>
<td>$R_{bd}=0$</td>
</tr>
<tr>
<td>$R_{bd'}$</td>
<td>$0.38 \times 10^{-4}$ ns$^{-1}$</td>
<td>$R_{bd'}=0$</td>
</tr>
</tbody>
</table>

As in the monoexciton case, we will calculate $R_{b0}$ and $R_{d0}$ from the electronic structure of the dot.

IV. CALCULATION OF BRIGHT AND DARK RECOMBINATION RATES FROM ELECTRONIC STRUCTURE

We use the empirical pseudopotential method where a superposition of screened atomic pseudopotentials is used to describe the quantum dot potential. We take spin-orbit interaction into account and the method naturally includes interband coupling and intervalley coupling. Following the diagonalization of the single-particle Hamiltonian, we use a configuration-interaction (CI) approach to obtain correlated monoexciton and biexciton wave functions $|\Psi^{(1)}(\chi)\rangle$ ($\chi =X^0, XX^0$). The characteristic radiative recombination rates $R_f (\chi)$ are calculated using Fermi’s golden rule from the correlated exciton wave functions as follows. For a transition $|\Psi^{(1)}(\chi)\rangle \rightarrow |\Psi^{(1)}(\chi-1)\rangle$, $R_f (\chi)$ follows from both the magnitude of the dipole matrix element of the transition $|\mathbf{M}_{f}^{(1)}(\chi)|^2$ and the recombination energy $\omega_f$. Namely,

$$R_f (\chi) = \frac{4G}{3} \left( \frac{e^2}{m_e c^2 \hbar^2} \right) n_0 \omega_f (\chi^n) \sum_{i=1,3,\pm 1} |\mathbf{M}_{f}^{(1)}(\chi)|^2.$$

Here, $e$ and $m_0$ are the charge and mass of the electron, respectively, and $c$ is the velocity of light in vacuum; the refractive index $\eta$ of the dot material accounts for the material’s effects on the photon emission; and $G = G(\epsilon_{\text{mat}}, \epsilon_{\text{sol}})$ accounts for the dielectric constant mismatch between the dot material ($\epsilon_{\text{mat}}$) and medium ($\epsilon_{\text{sol}}$)—solid barrier in self-assembled dots and liquid solvent in colloidal.

Table I shows the calculated characteristic radiative recombination rates $R_{b0}$ and $R_{b'} (i=b, b', d, d')$ for the monoexciton and biexciton, respectively, in a prototypical lens shaped In$_{0.4}$Ga$_{0.6}$As/GaAs self-assembled quantum dot with base diameter $b$= 252 Å and height $h$= 35 Å, and a colloidal CdSe quantum dot with diameter $D$=38 Å. We find that the rates for bright states indeed satisfy $R_{b0} = R_{b'/0}$ and that dark states obey $R_{d0} \sim R_{d'/0} \sim 0$.

V. ANALYTIC SOLUTION TO THE MODEL RATE EQUATIONS FOR THE MONOEXCITON

The calculated radiative rates for the In$_{0.4}$Ga$_{0.6}$As/GaAs self-assembled dot and the CdSe colloidal dot (Table I) show that it is a good approximation to consider $R_{d0}=0$. In this case, together with the assumptions that $R_{BD}=R_{DB}$ and $n_{b}(0)=n_{b'}(0)=1/2$, the solution of the model three-level system of rate equations [Eq. (6)] gives the following (the general framework for the analytic results is in the Appendix):

$$n_{b}(t) = F \exp(-\gamma_{b} t) + S \exp(-\gamma_{S} t)$$

with

$$\gamma_{b} = \frac{1}{2} (R_{b0} + 4R_{BD}) + \frac{1}{2} \sqrt{R_{b0}^2 + (4R_{BD})^2},$$

$$\gamma_{S} = \frac{1}{2} (R_{b0} + 4R_{DB}) - \frac{1}{2} \sqrt{R_{b0}^2 + (4R_{DB})^2}$$

and
In time-resolved PL experiments the measured signal \( I(t) \) is proportional to the number of photons per unit of time: \( dn_0/dt \) [Eq. (6)], which under the assumption of \( R_{BD} = 0 \) results in \( I(t) = R_{BD} n_B(t) \). Note that by being proportional to the occupation of the bright state the signal \( I(t) \) carries information on both radiative and nonradiative (spin flip) processes. Figure 2 shows the logarithm of \( n_B(t) \) as a function of time for different spin-flip rates \( R_{BD} \), and Fig. 3 shows the slow \((\gamma_S)\) and fast \((\gamma_F)\) components of the decay of \( n_B(t) \) versus \( R_{RD} \). We find that in the limiting cases of (1) extremely slow \( R_{BD} \ll R_{BD} \) and (2) extremely fast \( R_{BD} \gg R_{BD} \) spin flip the decay of \( n_B \) is primarily determined by a single exponential. In case (1) we find \( \gamma_F \approx 0 + 2R_{BD} \) and \( \gamma_S \approx 2R_{BD} \), while \( F = 1/2 \) and \( S \approx 0 \) (Fig. 3, inset). Here, \( \gamma_F \) resembles the expected result for the decay rate of the PL in the presence of nonradiative recombination centers; the dark state of the monoexciton in this case.\(^{10,11}\) The population of the bright state is

\[
  n_B(t) \approx \frac{1}{2} \exp\left[-\left(R_{BD} + 2R_{BD}\right)t\right].
\]  

In this regime, \( I(t) \) decays approximately with the characteristic lifetime of the bright states. In case (2), we find \( \gamma_F \approx 0 + 2R_{BD} \) and \( \gamma_S \approx 2R_{BD} \), and \( S \approx 1/2 \) and \( F \approx 0 \) (Fig. 3, inset); therefore,

\[
  I(t) = \frac{1}{2} \exp\left(-\frac{R_{BD}}{2} t\right).
\]

VI. COMPARISON WITH EXISTING EXPERIMENTAL DATA

In the experimental literature, data on the bright-dark transition time are scarce. \((In,Ga)As/GaAs \) dots. Dalginaro and co-workers\(^9,12\) have studied recently the effect of the dark state in the decay of the monoexciton in a gated structure and have estimated \( R_{BD} \approx 20 \) ns at a temperature of 5 K. In addition, they have found that this spin-flip rate varies strongly with the applied bias. From time-resolved PL experiments, Favero et al.\(^{13}\) have extracted significantly disparate values for two different dots: \( R_{BD} \approx 440 \) and 30 ns.

\( InP/(In,Ga)P \) dots. In a two-photon absorption experiment, Snoke and co-workers\(^9\) populated the dark states of the monoexciton and measured the subsequent luminescence as a function of time. From the data below 70 K, the authors found that the spin-flip, bright-dark transition time satisfies \( R_{BD} \approx 200 \) ps.

\( CdSe \) dots. By performing fluorescence transient experiments at room temperature, Wang et al.\(^{20}\) have concluded that the bright-dark, spin-flip relaxation times \( R_{BD} \approx 0.2 \)–0.4 ps. Thus, the spin-flip process is orders of magnitude faster than the characteristic radiative recombination time.

These experiments reveal order-of-magnitude variations in the bright-dark spin-flip times, suggesting that the value of \( R_{BD} \) appropriate to interpret time-resolved photoluminescence experiments in quantum dots is controversial and further research is needed to understand the spin-flip mechanism.
In the slow spin flip regime, consistent with the findings of Dalgaro et al.\textsuperscript{9} and Favero et al.,\textsuperscript{13} our calculated value $\tau_R(X^0) = R_{BD}^{-1} = 1.1 \text{ ns}$ for the In$_{0.6}$Ga$_{0.4}$As/GaAs dot is in excellent agreement with the data of Bardot et al.,\textsuperscript{14} who extracted 1.55 ns from time-resolved photoluminescence, and the value of 1 ns found by Buckle et al.\textsuperscript{15} and Stevenson et al.\textsuperscript{16} In the fast spin flip regime, for our prototypical CdSe dot (see Table I), we obtain $\tau_R(X^0) = 17 \text{ ns}$, which is in excellent agreement with the values of 17 and 19 ns extracted, respectively, by Brokmann et al.\textsuperscript{17} and Labeau et al.\textsuperscript{18} from time-resolved photoluminescence in ZnS-passivated CdSe dots. In the regime of intermediate spin flip rates, measurements of the biexponential decay of $I(t)$—as those performed by Dalgaro et al.\textsuperscript{9}—could be used to deduce the spin-flip rate.

**VII. THE RATIO $\tau_R(X^0)/\tau_R(XX^0)$**

To compare the biexciton decay rate with the monoexciton decay rate in the limiting cases discussed above (Sec. V), we first note that atomistic pseudopotential-based calculations show (Table I) that the characteristic radiative rates for the biexciton ground state satisfy

$$R_{0B}(XX^0) = R_{BD}(X^0),$$

$$R_{0D}(XX^0) = R_{DD}(X^0).$$

(16)

Second, in contrast to $X^0$, we note that the solution of the rate equation for $XX^0$ [Eq. (7)] results in $n_R(XX^0) \sim \exp(-\gamma t)$; a single exponential that decays with rate

$$\gamma = 2(R_{0B} + R_{0D}) = 2(R_{BD} + R_{DD})$$

(17)

regardless of the value of the spin-flip rate $R_{BD}$. Similarly to $X^0$, the time-resolved PL signal is proportional to the population of the bright state of the biexciton. For slow spin flip [case (1), Sec. V] we find a decay-rate ratio between $X^0$ and $XX^0$ of

$$\gamma' / \gamma_{X^0} = \frac{2R_{0B}}{R_{BD} + 2R_{DD}} \approx 2 \quad \text{(slow spin flip)},$$

(18)

and for fast spin flip [case (2), Sec. V] we find

$$\gamma' / \gamma_{XX^0} = \frac{2R_{0B}}{R_{BD}/2} \approx 4 \quad \text{(fast spin flip)}.$$  

(19)

We emphasize that depending on the magnitude of the spin-flip rate the ratio $\tau_R(X^0)/\tau_R(XX^0)$ can change by a factor of 2, therefore the assumed spin-flip time is crucial when comparing results for $\tau_R(X^0)/\tau_R(XX^0)$. Recently, Wimmer et al.\textsuperscript{7} have used a quantum Monte Carlo (QMC) approach with model single-band effective-mass electron and hole states to calculate

$$\tau_R(X^0)/\tau_R(XX^0) = 2 \quad \text{(slow spin flip)}.$$  

(20)

Those authors speculated that the disagreement with the pseudopotential and CI calculations of Ref. 8, which adopt the fast spin-flip regime and predict $\tau_R(X^0)/\tau_R(XX^0) = 4$ (fast spin flip), originates from an inaccurate treatment of correlations in CI. However, as is obvious from Eqs. (18) and (19), the discrepancy can be directly attributed to the different assumptions for the spin flip rates. When the same spin-flip rates are assumed, our results for $\tau_R(X^0)/\tau_R(XX^0)$ are in agreement with the QMC results of Wimmer et al.,\textsuperscript{7} suggesting that our treatment of correlations is adequate to predict the ratio of monoexciton and biexciton radiative lifetimes. On the other hand, the calculated values of the radiative recombination times depend not only on the treatment of correlation effects, but also on the accuracy of the electron and hole wavefunctions. Our atomistic results are in good agreement with experiment while the results of Wimmer et al.\textsuperscript{7} based on the single-band effective mass approximation differ from experimental data by a factor of 2.

Finally, note that in our calculation of $\tau_R(X^0)/\tau_R(XX^0)$ we assume that the change in occupation of the monoexciton bright and dark states [Eq. (1)] is not affected by the decay of the biexciton state.

**VIII. SUMMARY**

We calculated the characteristic radiative recombination rates for the ground state of the monoexciton and biexciton in self-assembled (In,Ga)As/GaAs and colloidal CdSe quantum dots using atomistic wave functions. For the monoexciton we used these rates in a model three-level system of rate equations where we varied the spin-flip rate $R_{BD}$. The latter affects significantly the radiative decay time: Fast spin flip leads to an exciton radiative recombination rate twice as fast as the rate obtained from slow spin flip. The radiative decay times $\tau_R(X^0)$ calculated in the limit of slow spin flip are in excellent agreement with available data for self-assembled dots, while for colloidal dots the agreement is best for fast spin flip. The biexciton radiative decay is a single exponential with a relaxation time that is independent of the spin-flip rate. But the ratio between the radiative decay time of the biexciton $\tau_R(XX^0)$ and monoexciton does depend on $R_{BD}$ and results, respectively, in $\tau_R(XX^0) / \tau_R(X^0) = 4$ and 2 for fast and slow spin flip. This result resolved the apparent contradiction between the calculation of Wimmer et al.,\textsuperscript{7} who predicted $\tau_R(XX^0) / \tau_R(X^0) = 2$, and our previous atomistic calculation\textsuperscript{8} in which we found $\tau_R(X^0) / \tau_R(XX^0) = 4$.

**ACKNOWLEDGMENTS**

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**APPENDIX: SOLUTION OF THE THREE-LEVEL MODEL RATE EQUATIONS FOR THE RADIATIVE DECAY OF $X^0$**

We use the simplified model of Eq. (6) as shown in Fig. 1(b). To find $n_R(t)$ and $n_D(t)$, we propose...
\[ n_D(t) = F \exp(-\gamma_D t) + S \exp(-\gamma_S t), \quad (A1) \]

with \( n_D(0) = F + S \) and \( \gamma_D \neq \gamma_S \), and solve first for \( n_D(t) \) with initial condition \( n_D(0) = 1 \). Then, to solve for \( S, F, \gamma_S \), and \( \gamma_D \), we substitute the solution of \( n_D(t) \) in the rate equation for \( dn_D/dt \) [Eq. (6)], obtaining the following conditions:

\[ \gamma_F^2 - (R_{BD} + 2R_{DB} + R_{D0} + 2R_{DB}) \gamma_F + R_{BD}R_{D0} + 2R_{DB}R_{D0} + 2R_{BD}R_{D0} = 0, \quad (A2) \]

Thus, we find

\[ \gamma_F = \frac{1}{2} \left[ (R_{BD} + 2R_{DB} + R_{D0}) + \frac{1}{2} \sqrt{(R_{BD} - 2R_{DB})^2 + 4(R_{BD} - R_{D0})(R_{BD} - R_{DB}) + 4(R_{BD} + R_{DB})^2} \right], \quad (A5) \]

\[ \gamma_S = \frac{1}{2} \left[ (R_{BD} + 2R_{DB} + R_{D0}) - \frac{1}{2} \sqrt{(R_{BD} - 2R_{DB})^2 + 4(R_{BD} - R_{D0})(R_{BD} - R_{DB}) + 4(R_{BD} + R_{DB})^2} \right], \quad (A6) \]

and

\[ F = \frac{R_{BD} + 2R_{DB} - \gamma_S}{\gamma_F - \gamma_S} n_D(0), \quad (A7) \]

\[ S = \frac{R_{BD} + 2R_{DB} - \gamma_F}{\gamma_F - \gamma_S} n_D(0). \quad (A8) \]

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*Present address: Eclipse Energy Systems, Inc., St. Petersburg, Florida 33710, USA.
Electronic address: gnarvaez@eclipsethinfilms.com


