Two Dimensional Förster Resonant Energy Transfer in a Mixed Quantum Dot Monolayer: Experiment and Theory

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ABSTRACT Energy transfer in monolayers of CdTe quantum dots (QDs), comprised of QDs of two different sizes, has been investigated. Förster resonant energy transfer (FRET) from QD donors to QD acceptors was observed and characterized using absorption and photoluminescence spectroscopy, as well as time-resolved photoluminescence measurements. A theory of FRET in two dimensions, taking into account random donor and acceptor distributions and exclusion zones around the donors, was used to fit the donor decays in mixed films. In this way the value of the Förster radius calculated from the spectral overlap was confirmed and a method for determining all important FRET parameters for QD monolayers from time-resolved photoluminescence measurements alone was found. The dependence of the measured FRET efficiency on the acceptor concentration can also be explained within this theory. Additionally, the rise in the acceptor emission enhancement for low acceptor:donor ratios indicates that energy is not only transferred from nearest neighbours but that donors at larger distances also contribute.

Introduction

Förster resonant energy transfer (FRET) is an energy transport mechanism based on dipole-dipole interactions[1,2]. Examples can be found in naturally occurring systems such as plants and bacteria in which energy absorbed from the sunlight is transported via FRET to reaction centers in the inner part of the organism to promote photosynthesis[3]. FRET can also be engineered in artificial structures in order to create light harvesting devices[3,4] and highly efficient nanosensors[5,6]. The use of semiconductor quantum dots (QDs) as energy donors and / or acceptors is of interest due to their photostability and high quantum yield, but especially because of their unique and tunable optical properties, providing a broad absorption range and a narrow emission feature[7,8]. FRET in pure QD systems, mainly three-dimensional mixed QD solids[9,10] and graded multilayer structures[11,12,13], has previously been reported. Highly efficient and fast energy transfer, with a transfer rate as high as $(50 \text{ ps})^{-1}$, was demonstrated by Franzl et al.[14]. This group also presented results on an energy funneling structure prepared from four different types of QDs, improving the emission from the central layer of QDs by a factor of four compared to a similar structure with seven layers containing the central-type of QDs only[4]. Understanding and improving FRET in a QD system is important in order to efficiently incorporate this mechanism in sensing devices or energy funneling structures.

The analysis of FRET in QD systems can however prove very complex as theoretical developments have mainly focused on molecular systems and it is arguable whether these can be directly applied to QD structures in the same way[12,15]. Despite the inhomogeneous broadening present in QD ensembles, the QD size and optical selection rules that have to be taken into account, it is at least possible to apply similar rate equations to describe FRET in QD structures[15]. FRET is typically characterised by two key parameters, firstly, the efficiency of the energy transfer mechanism and secondly, the Förster radius. The Förster radius represents the acceptor-donor separation at which the FRET efficiency reaches 50%. In molecular systems the Förster radius can be calculated based on the individual spectral properties of the donors and acceptors. However, in the case of inhomogenously broadened QD donor and acceptor ensembles it is not clear that the evaluation of the Förster distance based on their individual emission and absorption spectra is an accurate reflection of the real energy transfer distance within a mixed

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ensemble[12,15]. Additionally, in a mixed QD ensemble with a random distribution of donor-acceptor separations it is not possible to extract the FRET efficiency from these spectral measurements. Separate time-resolved measurements are necessary, often requiring a different experimental setup, which typically changes the illumination spot size and pumping and collection conditions. Thus, the determination of the FRET parameters and therefore, the characterization of FRET in a mixed QD structure is not straightforward. An analytical technique by which all the key FRET parameters could be extracted from a single measurement would be more robust and reliable.

We report on the optical properties of a monolayer structure prepared from mixed QD donors and acceptors using a layer-by-layer technique and compare our results with a theory of FRET in two dimensions previously only applied to biological systems. This theory allows for the extraction and therefore, verification of important parameters characterizing FRET such as the Förster radius, the FRET efficiency and the exclusion zone radius around the donors from lifetime measurements alone. The observed dependence of the FRET efficiency on the acceptor:donor ratio can also be explained within this theory. Additionally, we observe an increase in the acceptor emission enhancement per quantum dot with decreasing acceptor:donor ratio. This indicates that not only nearest neighbour donors transfer energy to a specific acceptor but donors at longer distances also contribute.

Experimental Methods

Two different sizes of negatively charged CdTe quantum dots (QDs), stabilized by thioglycolic acid[16], were deposited in pure and mixed monolayers on quartz slides. The 1.9 nm diameter QDs, emitting at 524 nm, were used as energy donors and the 3.5 nm diameter QDs, emitting at 617 nm, acted as energy acceptors.

The samples were prepared by the layer-by-layer (LBL) electrostatic assembly technique[17,18]. The pure or mixed QD monolayer was deposited directly on top of two polyelectrolyte bi-layers (each bi-layer consisting of a negatively charged poly(sodium 4-styrene sulfonate) and a positively charged poly(diallyldimethylammonium chloride) layer) on a quartz substrate. Further details on the sample preparation can be found elsewhere[19].

Samples with either a pure or a mixed monolayer of QDs were prepared by immersion in aqueous QD solutions containing either one type or two types of QDs. The concentrations in the pure films were varied by immersion of the substrates in solutions with different QD concentrations (between 0.5 and $2x10^{-6}$ M) for times varying between 1 and 20 minutes. The mixed QD layers with different acceptor:donor ratios were prepared by immersion in solutions containing varying ratios of acceptor to donor QDs. For samples with acceptor:donor ratios below 1:3 the donor concentration ($2x10^{-6}$ M) in solution was kept constant while adding acceptors. Alternatively to achieve samples with higher acceptor:donor ratios, donors were added to a solution with $2x10^{-6}$ M acceptors. The immersion time for the deposition of the mixed layers was kept constant at 20 minutes.

Absorption spectra of the films were recorded between 350 nm and 800 nm with a double beam UV-Vis Recording Spectrometer (Shimadzu UV-2401 PC). Room temperature photoluminescence (PL) spectra were recorded, with the excitation wavelength of 400 nm, using a Perkin-Elmer LS 55 fluorescence spectrometer. The time-resolved PL decays were measured using a PicoQuant Microtime200 time-resolved confocal microscope system with 150 ps resolution. Excitation was provided by 470 nm picosecond pulses at a 10 MHz repetition rate from a LDH-480 laser head controlled by a PDL-800B driver (PicoQuant). The PL decays were measured over an area of 80 µm by 80 µm (150 by 150 pixels) with an integration time of 4 ms per pixel. Broad band filters at 500 and 600 nm were used in the PL lifetime measurements to collect the emission from the donors and acceptors QDs respectively.

Results and Discussion

The properties of the pure QD layers are presented first, followed by the analysis of energy transfer and extraction of the FRET parameters in a mixed QD monolayer. Finally, the dependence of the FRET efficiency on the acceptor:donor ratio is discussed.

Figure 1 shows the PL (dashed lines, left-hand axis) and absorption spectra (solid lines, right-hand axis) of pure monolayers prepared from both types of QDs. The donor QDs have a diameter of approximately 1.9 nm and emit at 524 nm. For the donor data presented in figure 1 the concentration of

QDs in the layer is $(3.7\pm0.3)\times10^{17}$ m^{-2}. Both parameters, size and concentration of the QDs, can be extracted from the position of the first absorption peak and the absolute value at this wavelength[20]. The acceptor QDs have a diameter of approximately 3.5 nm and the emission from a monolayer with a concentration of $(1.1\pm0.2)\times10^{17}$ m^{-2} is centred at 617 nm. The acceptor QD peak emission wavelength and PL lifetime in a monolayer were found to be strongly dependent on the QD concentration in the layer. However, there was little concentration dependence observed for the donor QDs. These concentration dependent changes for the acceptor QD layers arise from energy transfer within the acceptor QD ensemble due to a comparatively large size distribution, reflected in a broader PL peak, and a small Stoke's shift[10-12]. The quantum yield of the QDs in the layer was determined by comparison to the luminescence standard Rhodamine 6G with a quantum yield of 95%[21]. The smaller donor QDs show a quantum yield of 4% in a layer, whereas the quantum yield for the acceptor QDs in a layer is only 1%.

As discussed above, one of the important parameters characterizing FRET is the Förster radius, R_0, representing the donor-acceptor separation at which the FRET efficiency is 50%. This distance can be calculated via the spectral overlap $J = \int_0^\infty \hat{I}_D(\lambda) \cdot \varepsilon_A(\lambda) \cdot \lambda^4 d\lambda$ where $\hat{I}_D(\lambda)$ is the area normalized donor emission spectrum and $\varepsilon_A(\lambda)$ the acceptor extinction spectrum, which can be obtained from the absorption spectrum via Lambert-Beers-Law[20]. The Förster radius

$$R_0 = 0.211 \cdot \left(\frac{\kappa^2 \cdot Q_D}{n^4} \cdot J \right)^{\frac{1}{6}}$$

also depends on the orientation factor κ (with $\kappa^2 = 2/3$ for randomly oriented dipoles as it is the case in the mixed monolayers investigated here), the donor quantum yield, $Q_D = (4\pm2)\%$, and the refractive index of the surrounding medium[2]. For the calculation of R_0 a volume weighted refractive index $n = 2.1\pm0.3$ was used. Based on the errors estimated for the determination of the spectral overlap, the measured donor quantum yield in a monolayer and the refractive index, a Förster radius of $R_0 = (3.3\pm0.2)$ nm was calculated for these donor and acceptor QDs.

The PL spectrum of a mixed QD monolayer, with donor and acceptor concentrations of (2.6±0.2) and (0.6±0.1)x10^{17} m^{-2} respectively, is shown in figure 2. For comparison the PL spectra of pure donor and acceptor layers with similar QD concentrations are also shown. Small differences in concentration were taken account for by weighting the PL spectra according to the concentrations extracted from the absorption spectra. The concentrations of the QDs in the mixed film were determined by fitting the mixed film absorption spectrum by a weighted sum of the pure acceptor and donor monolayer absorption spectra, which were shown in figure 1. A clear quenching of the donor emission and an increase of the acceptor emission can be observed in the mixed QD film. These are typical signatures for energy transfer and an FRET efficiency, E_{FRET} = (77±11)%, is estimated from the comparison of the integrated donor intensity in the pure and mixed layers. The acceptor emission in this monolayer with an acceptor:donor ratio of 1:4.5 increases only by a factor of 0.31±0.08.

Another important characteristic of FRET is the decrease of the average donor lifetime in presence of an acceptor, since the energy transfer mechanism presents an additional decay path for the excitons generated in the donors. As the absolute emission of the QD layers strongly depends on the QD concentration, it is more reliable to extract the energy transfer efficiency from the decrease in the average donor lifetime, as only small variations of the donor lifetime in a pure monolayer have been observed over the range of concentrations investigated here. The time-resolved photoluminescence measurements of the donors in a pure monolayer (filled circles) and a mixed monolayer (open symbols), with an acceptor:donor ratio of 1:4.5, are shown in Figure 3. The average lifetime is calculated using a two-exponential fit for both samples and decreases from τ_D=(6.1±0.3) ns in the pure layer to τ_{DA}=(2.5±0.3) ns in a mixed monolayer, corresponding to a FRET efficiency, $E_{FRET} = 1 - \dfrac{\tau_{DA}}{\tau_D}$, of (59±8)% and a FRET rate of (1.8±0.3 ns)$^{-1}$ for this acceptor:donor ratio.

For single donor-acceptor pairs and assemblies composed of a known acceptor:donor ratio it is possible to estimate the FRET efficiency $E_{FRET} = \dfrac{1}{1 + \left(\dfrac{R_{DA}}{R_0}\right)^6}$ from R_0 for a fix donor-acceptor

separation R_{DA}. However, in the case of a mixed QD monolayer as presented here, the random acceptor / donor distribution on formation of the layer results in a distribution of donor-acceptor separations. Therefore, a FRET efficiency cannot be estimated from the value of R_0 extracted from the spectral data of the pure monolayers and the time-resolved measurements on the mixed layer cannot provide an independent confirmation of the value of R_0. As described above, the calculation of R_0 is based on a number of experimentally determined parameters and assumptions. Independent verification of R_0 would confirm, for example, the validity of taking into account a volume weighted refractive index, as well as calculating the overlap from the full extinction spectrum opposed to taking into account the first absorption maximum only. To address these issues a theory of FRET between randomly distributed donors and acceptors in two dimensions[22] was investigated, as a mean of establishing an independent way of extracting the Förster radius from the FRET induced change in the donor lifetime.

Analogous to the earlier work by Förster in three dimensions[1], Wolber and Hudson[22] developed a theory to evaluate FRET in two-dimensional biological systems, such as lipid bilayer membranes. Along with the reduction from three to two dimensions this theory offers the possibility of taking into account exclusion zones around the donor. Due to the relatively large size of the QDs the interacting dipoles cannot come arbitrarily close to each other in the two dimensional monolayer structure investigated here. The minimum distance between the QD centres, and therefore the dipoles involved in the energy transfer, is determined by the QD radius as well as the length of the thioglycolic acid stabilizer of approximately 0.5 nm[23]. Taking into account the length of the stabilizer only once, as it was found that the ligand shells can interpenetrate[23,24], an exclusion zone with a radius $R_{ex} = (3.2\pm0.3)$ nm is created around the donor. According to the theory the donor decay depends on the average exciton lifetime τ_D in the pure film, the acceptor concentration c_{acc}, R_0, and R_{ex}. The dependence of the donor emission $I_{DA}(t)$ on time

$$I_{DA}(t) = \exp\left\{-\frac{t}{\tau_D} - \pi \cdot R_0^2 \cdot c_{acc} \cdot \gamma\left(2/3, (R_0/R_{ex})^6 \cdot (t/\tau_D)\right) \cdot \left(\frac{t}{\tau_D}\right)^{1/3} + \pi \cdot R_{ex}^2 \cdot c_{acc} \cdot \left[1 - e^{-(R_0/R_{ex})^6 \cdot (t/\tau_D)}\right]\right\}$$

is then described by an exponential decay based on the pure donor decay, characterized by the lifetime τ_D, and accelerated due to the energy transfer. The effect of the energy transfer is reduced due to the

lower limit of the donor-acceptor separation determined by the exclusion zone. As a multi-exponential decay is used to fit the pure donor decay (solid line in Figure 3), each exponent is modified according to the theory in order to fit the donor decay as measured in the mixed QD film using c_{acc}, R_0, and R_{ex} as the fitting parameters. As can be seen in figure 3, an excellent agreement with the data was achieved for c_{acc} = (0.75±0.1)×10^{17} m^{-2}, R_0 = (3.6±0.3) nm, and R_{ex} = (2.9±0.2) nm. These values agree well with those previously determined for these parameters, validating any assumptions made in the calculations. Fitting of the decays measured for mixed monolayers with different acceptor:donor ratios further confirmed these values of R_0 and R_{ex}. This shows that the theory presented by Wolber and Hudson cannot only be applied to molecular systems but can also be used to interpret FRET in two dimensional QD structures. Additionally, it allows for the extraction of all the important FRET parameters such as Förster radius, FRET efficiency and rate from time-resolved photoluminescence measurements alone.

Numerically integrating $I_{DA}(t)$ provides an expression for the FRET efficiency as a function of the acceptor concentration and Wolber and Hudson give an approximate equation for

$$E_{FRET} = 1 - \left(A_1\, e^{-k_1 C} + A_2\, e^{-k_2 C} \right) \text{ with } C = R_0^{\,2}\, c_{acc}.$$

The A and k values can be found in table 1 of reference [22] for several discrete values of R_{ex} / R_0. In figure 4 the FRET efficiency is plotted for four values of R_{ex} / R_0 (0.7, 0.8, 0.9 and 1.0) taking R_0 = 3.6 nm. The trend of the experimental data follows most closely the R_{ex} / R_0 = 0.8 line, and a corresponding FRET efficiency of 70% can be extracted for the acceptor concentration of c_{acc} = (0.75±0.10)×10^{17} m^{-2}. The dependence of the FRET efficiency on the acceptor concentration will be discussed further below. Dewey and Hammes provide a simpler approximation for the determination of the FRET efficiency in a two-dimensional structure which also takes account of exclusion zones[25]. Using equations (23) and (24) in their paper[25], for the case of an infinite plane, a FRET efficiency of (67±7)% was estimated for the values of c_{acc}, R_0, and R_{ex} obtained from the fit of the donor decay in the mixed monolayer presented in figure 3. The values obtained from the theory are in agreement with that previously extracted from the ratio of the average donor lifetimes in the pure and mixed monolayer.

Figure 4 shows the FRET efficiency as a function of the acceptor:donor ratio. The experimental FRET efficiencies are calculated using the average donor lifetimes. As the acceptor concentration increases and donor concentration decreases, the FRET efficiency increases as more acceptors per donor are available. This is a consequence of the increased probability for each donor to transfer its energy. Transfer efficiencies of over 90%, corresponding to transfer rates as high as $(0.5 \text{ ns})^{-1}$, were obtained for acceptor QD concentrations greater than $1.5 \times 10^{17} \text{ m}^{-2}$ in the mixed layer, corresponding to a monolayer with an acceptor:donor ratio of 1:1.

The enhancement of the acceptor emission per QD as a function of the acceptor concentration is plotted in figure 5. The enhancement is determined from the time-integrated PL decay of the acceptors in mixed and pure films with similar acceptor concentrations. Comparison of figures 4 and 5 show that even though the FRET efficiency continues to increase for acceptor concentrations greater than 1.0×10^{17} m^{-2}, corresponding to an acceptor:donor ratio of 0.3, there is no enhancement of the acceptor emission. For high acceptor:donor ratios the probability of energy transfer is high, but the number of donors is sufficiently low that the effect of the energy transfer on the overall emission per acceptor QD becomes comparatively small. At low acceptor:donor ratios a plateau, i.e. a constant emission enhancement per acceptor, would be expected. In this case the acceptors are sufficiently separated so that energy from the surrounding donors can only be transferred to one specific acceptor and this acceptor does not share donors with any other acceptor. The data shows that the enhancement of the acceptor emission per QD continues to increase down to a ratio of 1:33, the lowest experimentally investigated. This indicates that the acceptor is not only receiving energy from nearest neighbour donors. A similar effect has been reported by Achermann et al.[11], who investigated energy transfer in a close-packed monodisperse QD film prepared by the Langmuir-Blodgett technique. They found that acceptors can receive energy from donors located in up to the third shell in a short-range hexagonally ordered structure. Assuming such a hexagonally ordered structure for the low acceptor:donor ratio mixed QD monolayer presented here, with a central acceptor QD surrounded by donor QDs, an acceptor:donor ratio of 1:33 would include

donors in the third shell from the central acceptor QD. This is consistent with the earlier reported observations[11].

Conclusions

FRET in a monolayer prepared from two different sized QDs has been analysed and efficiencies as high as 90%, corresponding to a FRET rate of $(0.5 \text{ ns})^{-1}$, have been observed. Interpretation of the donor decay within a theory of FRET in two dimensions, based on random donor and acceptor distributions and taking into account exclusion zones around the donors, provides a means of extracting all important FRET parameters from the donor decays alone. This approach avoids any ambiguities which can arise when comparing values extracted from data recorded using different experimental measurement systems, different sample areas and under different pumping and collection conditions. Furthermore, comparison of the Förster radius evaluated using the two dimensional FRET theory of Wolber and Hudson and that extracted using the more traditional method of analysing FRET has confirmed the validity of the assumptions made regarding the evaluation of the spectral overlap function. Additionally, the acceptor concentration dependence of FRET in a two-dimensional system can be explained within the scope of this theory. The dependence of the acceptor emission enhancement per QD in a mixed film, compared to the emission from a pure film of the same concentration, suggests that not only closest neighbour donors transfer energy to a specific acceptor but that also donors located at least up to three shells from the acceptor QD also contribute to its emission enhancement.

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Figure 1. Emission (dashed lines) and absorption spectra (solid lines) of the donor and acceptor QDs in pure monolayers on quartz substrates.

Figure 2. Emission spectra for pure donor and acceptor QD monolayers (shown in green and red respectively) and for a mixed QD monolayer (black) with similar donor and acceptor QD concentrations. Small variations in the concentrations are taken account of by weighting the PL spectra with factors calculated from comparison of the absorption spectra.

Figure 3. Time-resolved photoluminescence measurements of the donor QDs in a pure (filled circles) and mixed monolayer (empty circles) with an acceptor:donor ratio of 1:4.5. The decay in the pure film can be fitted with a three exponential decay (black solid line). The faster decay in the mixed film is fitted using the Wolber and Hudson theory of FRET in two dimensions taking into account exclusion zones around the donors.

Figure 4. Dependence of the FRET efficiency on the acceptor QD concentration. The QD concentration, determined from the absorption spectra, is within an error of 20 %. The experimentally determined efficiency (red circles) follows the trend predicted by theory (black lines) for a ratio R_{ex} / R_0 ~ 0.8. The corresponding number of acceptor QDs per donor QD for the mixed films is given on the top axis.

Figure 5. Dependence of the acceptor emission enhancement factor per acceptor QD on the acceptor concentration in a mixed QD monolayer. The emission enhancement per QD was calculated with respect to the acceptor emission in a pure monolayer with a similar acceptor concentration. Weighting factors were used to take account of small variations in concentration.

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