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Anisotropic emission of interface fluctuation quantum dots

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Abstract. We calculate energy levels, dipole moments and radiative broadening of interface fluctuation quantum dots. For optically allowed states, the dipole moment grows proportionally to the lateral quantum dot radius while the radiative broadening saturates towards the quantum well radiative broadening for large lateral quantum dot radii. This is accompanied by a change in the angular emission pattern, concentrating emission in forward and backward direction. Optically forbidden states do not couple to light propagating in the growth direction yet they may have a considerable radiative broadening due to spontaneous emission in other directions.

PACS. 78.67.Hc Quantum dots – 78.60.-b Other luminescence and radiative recombination – 78.70.-g Interactions of particles and radiation with matter

1 Introduction

Interface fluctuation quantum dots (IFQD's) are large monolayer islands naturally formed when growing structures with growth interruptions [1–3]. Their large dipole moments may make possible the observation of quantum entanglement with a quantum dot nanocavity [4,5] which has aroused a considerable amount of interest in them recently.

Experiments generally measure either dipole moment or radiative lifetime but rarely both, so a theoretical relation between them could be valuable. Although there have been calculations of the IFQD dipole moment before [5], to the best of our knowledge there have been no publications of calculations of both dipole moment and radiative lifetime. For small quantum dots where the dipole moment is independent of the direction of the light wave vector it couples to, the dependence of the radiative lifetime on the dipole moment is simply quadratic, becoming, however, more complicated for large quantum dots where the coupling strength varies with the emission direction. The emission finally evolves toward the strict directional emission in the growth direction in the quantum well limit, where again the radiative broadening is simply proportional to the square of the dipole moment for coupling to the light propagating in the growth direction. Though the basic physics is unchanged in the case of large interface quantum dots, one should note that a lifetime measurement provides access to an angle integrated dipole moment while a direct measurement of the dipole moment *via* transmission or absorption gives only the corresponding directional component.

Recently, we have clarified the relation between dipole moment and radiative lifetime in semiconductor quantum dots [6]; here, we present actual calculations of the two quantities for interface fluctuation quantum dots. We also address the question of the angular dependence of IFQD emission which occurs for large dots and marks the beginning of a smooth transition to the ideal quantum well case where emission is in the forward and backward directions only.

Here, IFQD's are assumed to be spherically symmetric in the xy-plane whereas in reality they may be elongated in the $[1\bar{1}0]$ direction [2]. Since relatively little is known about the in-plane shape of IFQD's, we have not attempted a more exact description.

2 Calculation of dipole moment and radiative lifetime

We calculate the dipole moment and radiative broadening of an interface fluctuation quantum dot of radius R_0. Since R_0 is usually much larger than the two-dimensional exciton Bohr radius ($\approx$ 6 nm in GaAs), we assume the electron-hole relative motion to be unaffected by the in-plane confinement. Our theory therefore cannot describe the regime of strongly confined IFQD's with quantum dot radii $<$ 10 nm where Andreani *et al.* [5] calculate an increase in oscillator strength with decreasing quantum dot radius by diagonalization of a single-particle Hamiltonian.

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However, for the estimated IFQD size of 40–60 nm [2,3] it is fully applicable and due to its simplicity is not restricted to intermediate dot sizes as is the case with any method involving a large number of localized base functions [5]. The method is thus well suited for larger dots and for approaching the quantum well limit.

The exciton is localized within a circular well centered on the origin. The barrier height is determined by the difference of the confinement energy induced by a monolayer fluctuation. For the z-direction, we assume confinement in an infinite well of width L_z. We thus write the electron-hole wave function as

$$\Psi(\mathbf{r}_e, \mathbf{r}_h) = \psi(\mathbf{R}_\parallel)\phi_{1s}(\mathbf{r}_\parallel)\zeta_z(z_e)\zeta_z(z_h), \tag{1}$$

taking into account only $1s$ states. $\mathbf{R}_\parallel$ and $\mathbf{r}_\parallel$ are the in-plane center of mass and relative motion coordinates, respectively,

$$\mathbf{R}_\parallel = \frac{m_e}{m_e + m_h}\mathbf{r}_{e,\parallel} + \frac{m_h}{m_e + m_h}\mathbf{r}_{h,\parallel}, \tag{2}$$

$$\mathbf{r}_\parallel = \mathbf{r}_{e,\parallel} - \mathbf{r}_{h,\parallel} \tag{3}$$

with electron (hole) mass m_e (m_h) and electron (hole) in-plane coordinate $\mathbf{r}_{e,\parallel}$ ($\mathbf{r}_{h,\parallel}$).

$$\zeta_z(z_{e/h}) = \sqrt{\frac{2}{L_z}}\cos\left(\frac{\pi z_{e/h}}{L_z}\right) \tag{4}$$

is the electron/hole wave function in z-direction. $\phi_{1s}(\mathbf{r}_\parallel)$ is the $1s$ exciton wave function and $\psi(\mathbf{R}_\parallel)$ is the center of mass wave function of the exciton. For a circular lateral quantum well and using polar coordinates in the xy-plane, the wave function becomes separable and we have

$$\psi(\mathbf{R}_\parallel) = R_n(R_\parallel)\frac{1}{\sqrt{2\pi}}\mathrm{e}^{\mathrm{i}m\phi}, \tag{5}$$

where n is the radial and m the angular quantum number. The radial wave functions $R_n(R_\parallel)$ are Bessel functions both inside and outside the dot which have to be matched together correctly at the well boundary.

The dipole moment can then be calculated as [9]

$$\begin{aligned}\mathbf{D}(\mathbf{K}) &= \mathbf{d}_{cv}\int \mathrm{d}^3R\,\Psi(\mathbf{R},\mathbf{R})\,\mathrm{e}^{-\mathrm{i}\mathbf{K}\cdot\mathbf{R}}\\ &= \mathbf{d}_{cv}\int \mathrm{d}^3r_e\int \mathrm{d}^3r_h\,\Psi(\mathbf{r}_e,\mathbf{r}_h)\,\mathrm{e}^{-\mathrm{i}\mathbf{K}\cdot\mathbf{r}_e}\delta(\mathbf{r}_e-\mathbf{r}_h)\\ &= \mathbf{d}_{cv}\phi_{1s}(\mathbf{r}_\parallel=0)\int \mathrm{d}^2R_\parallel\,\psi(\mathbf{R}_\parallel)\mathrm{e}^{-\mathrm{i}\mathbf{K}_\parallel\cdot\mathbf{R}_\parallel}\\ &\quad\times\int \mathrm{d}Z\,|\zeta_z(Z)|^2\mathrm{e}^{-\mathrm{i}K_zZ}.\end{aligned} \tag{6}$$

Inserting equation (5) into equation (6) and calculating the dipole moment for coupling with light propagating in the growth direction, $\mathbf{K} = K\,\mathbf{e}_z$ we immediately see that the dipole moment vanishes for all $m \neq 0$-states. These are usually called "optically forbidden" states. However, we will later show that they have a finite radiative broadening. Equation (6) also shows that for small quantum dots where $\mathrm{e}^{-\mathrm{i}\mathbf{K}\cdot\mathbf{R}}$ varies little over the quantum dot, the dipole moment becomes independent of the direction of the light wave vector $\mathbf{K}$.

The radiative broadening of a semiconductor quantum dot is [6]

$$\begin{aligned}\Gamma_{rad} = \frac{n\pi\omega^3}{(2\pi)^3\epsilon_0\hbar c_{vac}^3}\sum_{\sigma=1}^{2}\int_0^\pi \mathrm{d}\theta\sin(\theta)\\ \times\int_0^{2\pi}\mathrm{d}\phi\,|\mathbf{D}(K_0,\theta,\phi)\cdot\mathbf{e}_\sigma|^2.\end{aligned} \tag{7}$$

Here, $n = \sqrt{\epsilon}$ is the index of refraction of the semiconductor material and K_0 is the wave vector corresponding to the frequency ω, $K_0 = n\omega/c_{vac}$. $\mathbf{e}_\sigma$ is the polarization vector of the light with polarization σ. $\mathbf{D}(K_0,\theta,\phi)$ is the dipole moment for coupling to light with wave vector K_0 propagating in the direction given by the angles (θ,ϕ). We have assumed the same index of refraction for quantum dot and medium.

For small quantum dots where the dipole moment is independent of angles θ and ϕ, we immediately recover the well-known quadratic dependence of the radiative broadening on the dipole moment from equation (7),

$$\Gamma = \frac{n\omega^3|D|^2}{3\pi\epsilon_0\hbar c_{vac}^3}\,. \tag{8}$$

In the quantum well limit, the exciton center of mass wave function is a plane wave with wave vector $\mathbf{Q}_\parallel$, $\psi(\mathbf{R}_\parallel) = \mathrm{e}^{\mathrm{i}\mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel}/\sqrt{A}$ where A is the quantization area. Equation (6) thus reads

$$\begin{aligned}\mathbf{D}(\mathbf{K}) &= \mathbf{d}_{cv}\phi_{1s}(\mathbf{r}_\parallel=0)\int \mathrm{d}^2R_\parallel\frac{\mathrm{e}^{-\mathrm{i}(\mathbf{K}_\parallel-\mathbf{Q}_\parallel)\cdot\mathbf{R}_\parallel}}{\sqrt{A}}\\ &\quad\times\int \mathrm{d}Z|\zeta_z(Z)|^2\mathrm{e}^{-\mathrm{i}K_zZ}\\ &= \mathbf{d}_{cv}\phi_{1s}(\mathbf{r}_\parallel=0)\delta_{\mathbf{K}_\parallel,\mathbf{Q}_\parallel}\sqrt{A}\int \mathrm{d}Z|\zeta_z(Z)|^2\mathrm{e}^{-\mathrm{i}K_zZ},\end{aligned} \tag{9}$$

yielding constant oscillator strength per unit area. Assuming spatially homogeneous excitation in growth direction, only states with vanishing in-plane momentum $\mathbf{Q}_\parallel = 0$ are excited. The radiative broadening in a quantum well then becomes [10,11]

$$\Gamma_{rad} = \frac{\omega d_{cv}^2|\phi_{1s}(r=0)|^2}{nc_{vac}\epsilon_0\hbar}\,. \tag{10}$$

3 Results

We have first calculated the energy levels of exciton states in a GaAs/$\mathrm{Al}_x\mathrm{Ga}_{1-x}$As IFQD for quantum well widths of $L_z = 3$ nm, 6 nm and an Al content of the barrier $x = 0.3$,

Table 1. Lowest four states of an IFQD with a radius of 20 nm and different well width L_z and Al-contents of the barrier x ($GaAs/Al_xGa_{1-x}As$ structure). Energies are measured from the bottom of the quantum dot. The barrier heights in the in-plane direction, *i.e.* the differences in quantization energy induced by a monolayer fluctuation, are also given.

		Energy(meV)		
		$L_z = 3$ nm	$L_z = 6$ nm	
in-plane barrier		16.1 meV	5.4 meV	9.1 meV
n	m	$x = 0.3$	$x = 0.3$	$x = 1.0$
0	0	2.2	1.7	2.0
0	1	5.5	4.1	4.9
0	2	9.7	unbound	8.3
1	0	11.0	unbound	8.9

1.0 [12]. The lowest four energy levels are listed in Table 1 for a typical lateral quantum dot radius of 20 nm. We see that in the 6 nm $GaAs/Al_{0.3}Ga_{0.7}As$ quantum well, only two states are bound, of which only one, *i.e.* the ground state, is optically active. The variation in well widths and barrier Al content induces ground state energy changes of up to 0.5 meV.

We want to stress that the values for quantization energies as well as energy separation between levels are heavily dependent on the exact shape of the confining potential. This is especially important because IFQD's have been found to be elongated in the $[1\bar{1}0]$ direction [2]. We discuss the effects of elongation for the case of a rectangular quantum dot with infinite barriers and side lengths L_x, L_y. The ground state energy is then $E_G = \pi^2\hbar^2/(2ML_x^2) + \pi^2\hbar^2/(2ML_y^2)$ where M is the exciton mass, $M = m_e + m_h$. Keeping the area $A = L_xL_y$ constant, we find that E_G is minimal for a square and may grow to arbitrarily large values for an elongated rectangle. Considering the energy splitting between ground state and first excited optically active state, we find just the opposite: For $L_x \geq L_y$, the splitting is $\Delta E = 8\pi^2\hbar^2/(2ML_x^2)$. The energy separation thus has a maximum for a square shape and becomes arbitrarily small for an elongated rectangle.

From equation (6) we see that as long as interference effects due to the finite wavelength of light are negligible ($KR_0 \ll 1$), the dipole moment is largely dependent on the area covered by the center of mass exciton wave function $\psi(\mathbf{R}_\parallel)$ and not on the shape of the confining potential. This becomes important when comparing energy separations and dipole moments for states in different quantum dots. If the dots are of different shape, the values of energy separation and dipole moment may seem completely unrelated.

As mentioned before, IFQD's in the GaAs/AlGaAs system have been found to be elongated in the $[1\bar{1}0]$ direction. This shape asymmetry leads to a fine structure splitting of the emission lines which was found to be as large as 25 μeV for the ground state [2]. The fine structure splitting as a function of the elongation has been calculated by Ivchenko [13].

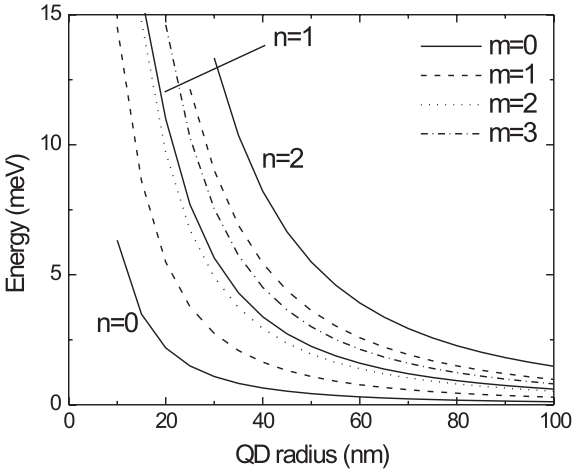

Fig. 1. Lowest seven energy levels of an interface fluctuation quantum dot depending on the dot radius. States are labelled according to their angular momentum m. Only $m = 0$-states are optically allowed ($GaAs/Al_{0.3}Ga_{0.7}As$ quantum well, $L_z = 3$ nm).

From hereon, all calculations are for the case of a well width of 3 nm and Al content of the barrier $x = 0.3$. Figure 1 shows the energy levels of the lowest seven IFQD states depending on the quantum dot radius. The solid lines mark the optically active states ($m = 0$) which may be observed in absorption experiments.

In Figure 2, we have plotted the dipole moments for coupling to light propagating in z-direction for the first three optically active states ($m = 0$, solid lines). We see that they rise almost linearly over the entire range of IFQD radii, reflecting their proportionality to the square root of the area covered by the wave function. The excited states show a deviation for the smallest calculated IFQD radii because the states are weakly bound and thus extend far out into the barrier.

An approximate relation between the dipole moments and thus oscillator strengths of different states can be found. Consider a circular quantum dot with infinite barriers in the in-plane direction and a radius R_0. The exciton center of mass wavefunction is then

$$\psi(R'_\parallel) = \begin{cases} \dfrac{1}{\sqrt{\pi}\mathrm{J}_1(k_n)}\mathrm{J}_0(k_nR'_\parallel),\ R'_\parallel < 1 \\ \\ 0, \qquad\qquad\qquad R'_\parallel > 1 \end{cases} \tag{11}$$

where $R'_\parallel = R_\parallel/R_0$, $\mathrm{J}_0(k)$ and $\mathrm{J}_1(k)$ are Bessel functions and k_n is a zero of the Bessel function $\mathrm{J}_0(k)$ ($k_0 = 2.4$, $k_1 = 5.5$, $k_2 = 8.7$). Neglecting the $\mathbf{K}$-dependence in equation (6), we calculate for small quantum dots ($K_0R_0 \ll 1$, *i.e.* the quantum dot has to be small compared to the wavelength of light)

$$D(\mathbf{K}) = \mathbf{d}_{cv}\phi_{1s}(\mathbf{r}_\parallel = 0)\frac{2\sqrt{\pi}R_0}{k_n}\,. \tag{12}$$

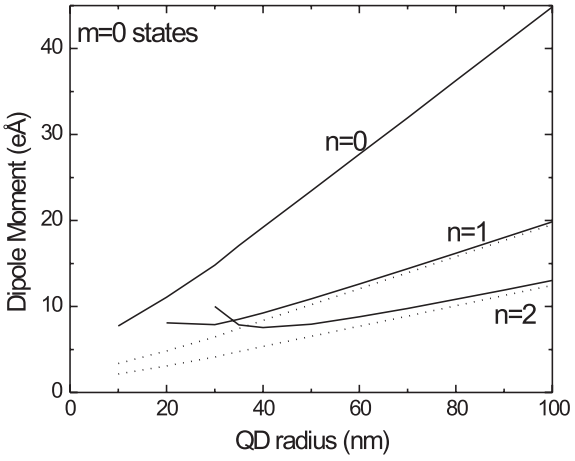

Fig. 2. Dipole moment for coupling to light propagating in the growth direction for the first three $m = 0$ states (solid lines); $m \neq 0$-states have a vanishing dipole moment. The dotted lines show approximations for the dipole moment of the excited states as discussed in the text (1 eÅ = 4.8 Debye, calculation for a GaAs/$Al_{0.3}Ga_{0.7}$As quantum well, $L_z = 3$ nm).

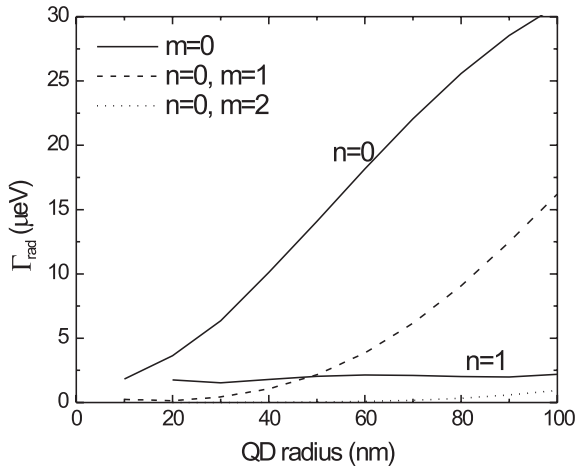

Fig. 3. Radiative broadening for the lowest four states. Even though the $m \neq 0$-states are optically forbidden, they may show a considerable radiative broadening (GaAs/$Al_{0.3}Ga_{0.7}$As quantum well, $L_z = 3$ nm).

Thus, we retrieve the linear dependence on the quantum dot radius and find that the ratio of the dipole moments of the two optically active states with quantum numbers (n,0) and (n',0) is $k_{n'}/k_n$. Combining the ground state dipole moment with the above ratio leads to a reasonable approximation for the higher states; see Figure 2, dotted lines. We find good agreement with the values calculated for a finite quantum dot except for the very weakly bound states where the IFQD wave function reaches out far into the finite barrier.

In Figure 3, we plot the radiative broadening ($\hbar$/lifetime) of the lowest four states. Although the $m \neq 0$-states do not couple to light propagating in the growth direction, they may still show a considerable radiative broadening; see the $m = 1, n = 0$-state (dashed line). This is due to their finite spontaneous emission in directions other than the growth direction. For small quantum dots ($KR_0 \ll 1$), spontaneous emission is almost direction-independent [6]. Accordingly, the radiative broadening for the $m \neq 0$-states is very small. Only when the dot size reaches the order of the transition wavelength in the semiconductor medium (*i.e.* $KR_0 \approx 1$) does the radiative broadening become significant because the finite radiative broadening of $m \neq 0$-states is essentially a consequence of interference effects between exciton wave function and light field. For small quantum dot radii, the radiative broadening of the first excited state ($n = 0, m = 1$) rises like R_0^3.

The radiative broadening of $m = 0$-states, by contrast, shows a very different dependence on the quantum dot radius. The ground state ($m = n = 0$) radiative broadening rises quadratically for small lateral radii while saturating towards the limit of quantum well radiative broadening for large lateral extensions. In a quantum dot with infinite barriers in the in-plane direction, every optically allowed state shows a quadratic rise of the radiative broadening for small IFQD radii. For finite barriers, however, higher optically allowed states are not bound for these radii. Also, their radiative lifetimes saturate at lower IFQD radii than the ground state. The saturation is due to interference effects between light field and exciton wave function and the fact that the wave function of an excited state varies faster than that of the ground state (n denotes the number of zeroes of the exciton wave function); this is easily understandable. Looking at Figure 3, note that the radiative broadening of the first optically active excited state ($n = 1, m = 0$) varies very little, *i.e.* it is saturated whenever it is bound for the parameters considered here.

We will now turn to the investigation of the direction dependence in IFQD's, considering the electron-heavy hole transition where the dipole moment lies in the in-plane direction. With nonresonant excitation in circular quantum dots, the emission intensity of an ensemble of quantum dots depends only on the azimuthal angle θ. Direction dependent emission intensity is governed by the scalar product of the dipole moment and the light polarization vector, $\mathbf{d}_{cv} \cdot \mathbf{e}_\sigma$, which introduces a factor $(1 + \cos^2\theta)/2$, and the absolute value of the dipole moment, $D(\mathbf{K})$. For small quantum dots, $D(\mathbf{K})$ is independent of the direction of the light wave vector $\mathbf{K}$. For larger sizes, $D(\mathbf{K})$ becomes more and more anisotropic and, for the case of the IFQD ground state, concentrated towards the z-direction. This culminates in the ideal quantum well limit ($R_0 \to \infty$) where emission is in $\pm z$-direction exclusively. In Figure 4 we plot the emission from the quantum dot ground state for different IFQD radii. The angle is measured with respect to the sample normal. The emission is normalized to the emission in the normal direction for each IFQD radius. We observe that for a radius of 10 nm, the emission varies by a factor of 2 due to the factor $(1 + \cos^2\theta)/2$, while for a 100 nm-quantum dot the

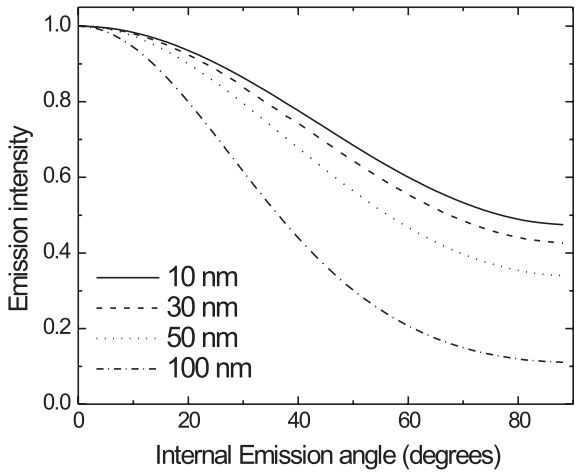

Fig. 4. Angle dependence of the emission of the ground state for different IFQD radii. The emission becomes more and more concentrated in forward and backward direction for larger radii, finally reaching the limit of a quantum well where emission is in the forward and backward directions only ($GaAs/Al_{0.3}Ga_{0.7}As$ quantum well, $L_z = 3$ nm). The emission is normalized to the emission in growth direction.

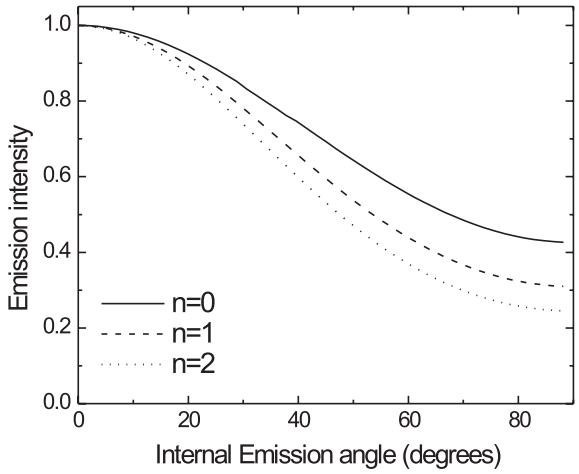

Fig. 5. Angle dependence of the emission of optically allowed states for an IFQD radius of 30 nm. Higher states show a higher degree of anisotropy at the same radius due to the faster variation of their wave functions ($GaAs/Al_{0.3}Ga_{0.7}As$ quantum well, $L_z = 3$ nm). The emission is normalized to the emission in growth direction.

emission in the in-plane direction is only 10% of the emission into the same solid angle in the growth direction. For even larger IFQD radii, the rate of emission (*i.e.* the radiative broadening) stays constant with growing dot radius but the emission cone becomes narrower. The degree of anisotropy depends on the value of the product of the quantum dot radius and the transition wave vector in the medium, $K_0 R_0$.

As mentioned above, higher excited states have faster varying wave functions than the ground state and thus show a higher degree of anisotropy at the same quantum dot radius. This is illustrated in Figure 5 where we plot the angle dependence of the first three optically allowed states for an IFQD radius of 30 nm. The 2nd excited state emission decreases by 75% when varying the emission angle from the normal direction to the in-plane direction, while the ground state emission in the in-plane direction is about 40% of the emission in the z-direction.

4 Conclusion

We have calculated the energy levels, dipole moments and radiative broadening of interface fluctuation quantum dots. The quantum dot states can be divided into optically allowed/optically forbidden states which couple/do not couple to light propagating in the growth direction. Optically forbidden states, however, can still be radiatively broadened if they have a finite dipole moment for light wave vectors in directions other than the growth direction. Their radiative broadening is very small for lateral IFQD radii which are small compared to the wavelength of light in the medium but may increase sharply for larger quantum dots.

The dipole moment of an optically allowed exciton state, by contrast, grows linearly with quantum dot radius. Consequently, the radiative broadening grows quadratically for small quantum dot sizes. Once the IFQD radius becomes of the order of magnitude of the wave length of light in the medium, however, the radiative broadening saturates and converges towards the quantum well value of 32 μeV (GaAs) for large quantum dots. This saturation is accompanied by a change of the angular emission pattern in the form of a smooth transition from the emission in all directions of a quantum dot to the emission into the forward and backward direction exclusively of a semiconductor quantum well. For higher excited states, saturation as well as anisotropy of emission occur at smaller dot radii than for the ground state.

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