Queen Mary, University of London

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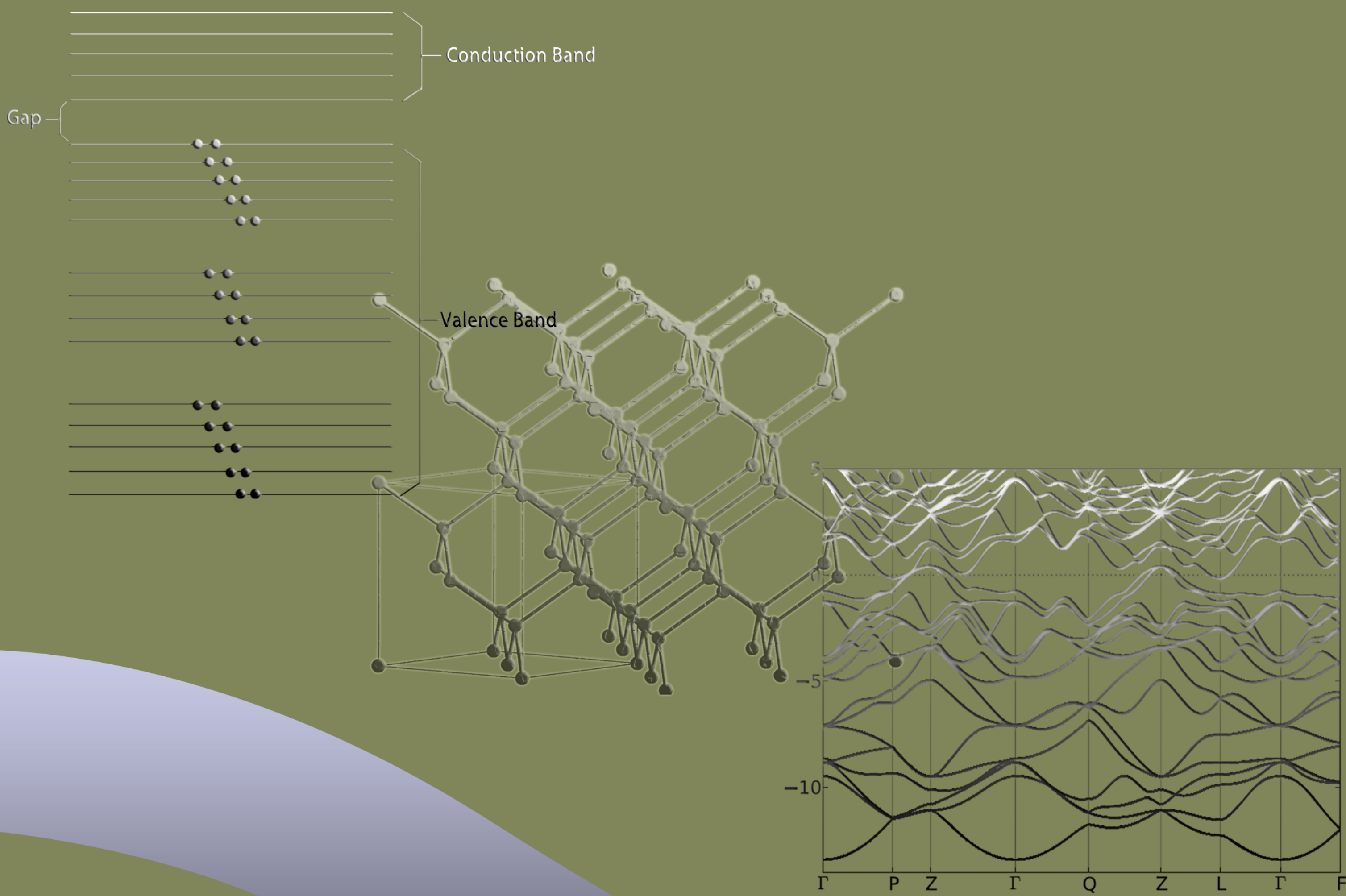
Photoluminescence Phenomenon in Multi-shell Quantum Dot



Discussion

For the multi-shell quantum dots (QDs), one of the most important and complex matter is optical properties, e.g. PL and PLE property (See Appendix). Especially for the light emission behavior, there are still debate on where the emission comes from (direct & indirect recombination, defects, etc.). This short material provides the discussion on contribution to light emission from the several layers (typically three layers, which is used for this material) of multi-shell QD, which favors the understanding of optical property of multi-shell QD.

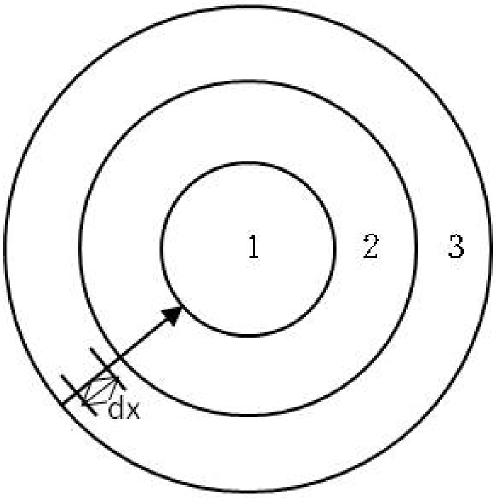


Figure 1. Model of three-shell quantum dot

Fig.1 shows the model for three-shell QD, briefly speaking, the incoming light is absorbed by either of the three layers, but the light emssion could come from both the direct recombination and indirect energy transfer between several layers. To discuss the optical property, it is necessary to introduce μ for absorbtion coeffient and η for light emission coeffient. Then we define the light intensity against distance x, namely I(x). The widely accepted expression is $I(x) = I_0 e^{-\mu x}$ (I_0 is the incoming light intensity). Through distance dx, the absorbed light intensity should be $\mathrm{d}I_{ab} = \mu I(x)\mathrm{d}x$, then the emission light intensity becomes $\mathrm{d}I_{LE} = \mu\eta I(x)\mathrm{d}x$. As is discussed above, for the multi-shell structure, the light absorbed by layer 1 can then excite light from layer 1, 2 and also 3. (the similar stuff occurs within each layer). So the emission expression becomes:

$$\mathrm{d}I_{LE} = I_0 \sum_{i,j=1}^{3} \mu_i \eta_j e^{-x\sum_i^3 \mu_i} \mathrm{d}x \tag{0-1}$$

then we can calculate the integration to obtain the emission intensity:

$$I_{LE} = I_0 \sum_{i,j=1}^{3} \mu_i \eta_j \left(\frac{1 - e^{-\sum_i^3 \mu_i d}}{\sum_i^3 \mu_i}\right) \tag{0-2}$$

By deducting the isolated atom absorbtion, the normalized absorbtion coefficient can be defined as: $\chi = \frac{\mu-\mu_0}{\mu_0}$, then $\mu = \mu_0(1+\chi)$. Replacing μ in expression 0-2, we can get:

$$\begin{aligned} I_{LE} &= I_0 \sum_{i,j=1}^{3} \mu_i \eta_j (1+\chi_i)(\frac{1-e^{-\mu_0 \sum_i^3 (1+\chi_i)d}}{\mu_0 \sum_i^3 (1+\chi_i)}) \\ &= \frac{I_0 \mu_0 \sum_{i,j}^3 \eta_j (1+\chi_i)(1-e^{-\mu_0 \sum_i^3 (1+\chi_i)d})}{\mu_0 \sum_{i,j=1}^3 (1+\chi_i)} \end{aligned} \quad (0\text{-}3)$$

The expression for I_{LE} here is hard for discussion, e.g. if we want to further discuss the contribution to light emission of the three layers, it is difficult to separate individual terms for each of the three layers directly from expression 0-3. Hereby, we might assume the isolated contribution from each of the three layers as well, which is demonstrated in Fig. 2.

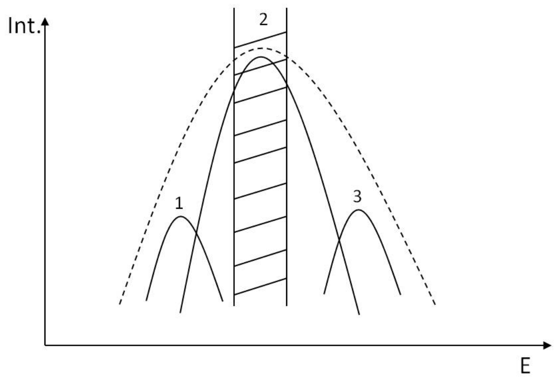


Figure 2. Assumption of separate contribution from each layer to PL

For Fig. 2, if we focus on layer 2, then it is easy to only take μ_2 into account based on the assumption, so expression for emission intensity then becomes:

$$I_{LE} = I_0 \eta_2 \frac{\sum_i^3 (1+\chi_i)}{\sum_i^3 (1+\chi_i)} (1-e^{\mu_0 \sum_i^3 (1+\chi_i)d}) \quad (0\text{-}4)$$

For expression 0-4, because the size d for QD is tiny (<10nm), we can only keep the first two terms for the Taylor expansion. Then we have:

$$\begin{aligned} I_{LE} &= I_0 \eta_2 \{1 - [1 - \mu_0(3+\chi_1+\chi_2+\chi_3)d]\} \\ &= dI_0 \mu_0 (3\eta_2 + \eta_2\chi_1 + \eta + 2\chi_2 + \eta_2\chi_3) \end{aligned} \quad (0\text{-}5)$$

From expression 0-5, we can discuss the light emission contribution of the three layers. The I_{LE} here only refers to layer 2 (see the illustraion for this assumption in Fig. 2), so if we want to believe the contribution for I_{LE}(actually, I_{LE} here refers to light emission only from layer 2) also only comes from layer 2, there shouldn't be the cross terms '$\eta_2\chi_1$' and '$\eta_2\chi_3$' in expression 0-5. This means the electron excited in layer2 does not go into layer 1 nor 3, which requires the electron mean free path is shorter than the size of layer 2. However, there is nearly not research carried out on the discussion of electron mean free path in QDs, which is obviously different from that in bulk materials.

Here, it is important to notice that the coefficient μ and η only concern the optical behavior. Although it is defined that $\chi = \frac{\mu-\mu_0}{\mu_0}$, which of course with close relationship to light absorbtion property, actually χ concerns more about electron motion in the material andthe interaction between electron and phonon. So it makes sense that we consider all cross terms for light behavior first and then separate χ_i (i=1,2,3,$\cdots$) from η_j (j=1,2,3,$\cdots$).

Appendix

PL (Phtoluminescence): the intensity response of emission light with different wavelength to the set excitation incoming wavelength.

PLE (Phtoluminescence Excitation): the response of the emission light intensity within set wavelength range to different excitation wavelength.