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RESEARCH ARTICLE

Temperature dependence of photoluminescence of QD arrays

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Abstract It is essentially important to understand the temperature dependence of the photoluminescence of multimodal quantum dot (QD) arrays for the realization of efficient photonic devices. In this paper, the dynamics processes of different density multimodal QD arrays were fitted by using the rate equation model. It is shown that, in high density QD arrays, the intensity of photoluminescence of different QD families has different temperature dependence, and the intensity of photoluminescence is quenched as the temperature increases in low density QD arrays. In high density QD arrays, as the temperature increases, the carriers will be thermally excited into the wetting layer from QDs, and then some of them will be recaptured by the big scale QDs; carrier coupling takes place between the different QD families, while in low density QD arrays, the carrier transfer between different QD families will be limited. Temperature dependence of the maximum of the ratio of photoluminescence intensity of different QD families strongly depends on the difference of thermal activation energies.

Keywords optoelectronics, rate equation, photoluminescence, multimodal quantum dot (QD) arrays, thermally excited

1 Introduction

Semiconductor quantum dots (QDs) material has very important theoretical value and broad application prospect [1–4]. We can research the structure and the physical characteristic of QDs by using different experimental methods, and it is essential to research the temperature dependence of the photoluminescence (PL) spectrum of the QDs to understand the operative characteristic of the QDs device. Further studies on the temperature dependence of the PL of the QD arrays have important significance to improve the characteristic of the quantum dots device.

The structure of the multimodal QD arrays consists of several different QD families (QDs size model). Different families have their own size and density [5]. Wetting layer plays an important role in the process of carrier transfer [6]. In a high density QD arrays system, the dynamics process exists in different families as follows: i) carriers' thermal escape from QD; ii) carriers are excited to the wetting layer and recaptured by QD, so the carrier transfer is realized in different QDs through the wetting layer [7]. Comparatively, the dynamic process in low density QDs is simple.

Temperature dependence of PL and PL thermal quenching phenomenon with different density QD arrays is simulated in this paper. Temperature dependence of the ratio of the PL intensity in multimodal QD arrays is also discussed. In multimodal QDs, the ratio of PL intensity in different QD families could reflect the distribution of the carriers in QDs. Based on the relationship between the temperature and the ratio of PL intensity, the rule that the distribution of carriers depend on temperature is researched.

2 Theoretical model

A model is shown in Fig. 1 to simulate the temperature dependence of the dynamic process in a multimodal QD arrays system, and suppose the following:

1) The generation rate in the potential barrier layer is g;
2) Omit the direct excitation or capture between the QDs and potential barrier layer;
3) The number of carriers in the wetting layer is m, thermal excitation rate is R_e', non-radiant recombination rate is R';
4) Suppose w QD families with different sizes, n_i is the number of carriers in different QDs, the carrier capture rate from the wetting layer is C_i, thermal excitation rate is R_{ei}, radiation combination is R_i, R_i has the same value in this model;

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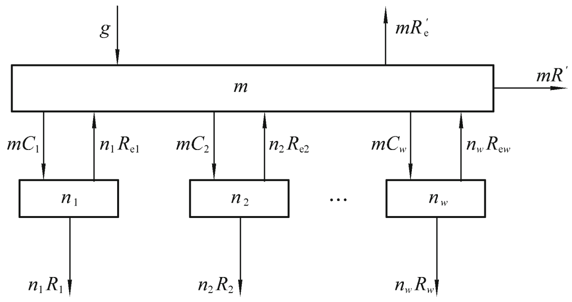

Fig. 1 Model of carrier dynamics processes in multimodal QD arrays

5) Omit the temperature dependence of parameters C_i, R_{ei}, R_i, R_e', g, R';

6) Consider only the QD ground state in the model, and the assumption is rational to low excitation densities.

The steady-state rate equation was built based on the above theoretical model

$$\frac{dm}{dt}=g-mR_e'-mR'-m\sum_{i=1}^{w}C_i+\sum_{i=1}^{w}n_iR_{ei}=0, \quad (1)$$

$$\frac{dn_i}{dt}=mC_i-n_iR_{ei}-n_iR_i=0. \quad (2)$$

From Eq. (2),

$$n_i=\frac{mC_i}{R_{ei}+R_i}. \quad (3)$$

Substituting Eq. (1) with Eq. (3), we obtain

$$m=g\left[\left(R_e'+R'\right)+\sum_{i=1}^{w}C_i-\sum_{i=1}^{w}\frac{C_iR_{ei}}{R_{ei}+R_i}\right]^{-1}. \quad (4)$$

By deduction, the PL intensity of QD radiation combination is

$$I_i=R_in_i=R_ig\left[\left(R_e'+R'\right)+\sum_{i=1}^{w}C_i-\sum_{i=1}^{w}\frac{C_iR_{ei}}{R_{ei}+R_i}\right]^{-1}\frac{C_i}{R_{ei}+R_i}. \quad (5)$$

If the dynamic process in bimodal and QD is considered arrays only, the equation of PL intensity in QD could be simplified as

$$I_i=R_in_i=R_ig\left[\left(R_e'+R'\right)+\sum_{i=1}^{2}C_i-\sum_{i=1}^{2}\frac{C_iR_{ei}}{R_{ei}+R_i}\right]^{-1}\frac{C_i}{R_{ei}+R_i}. \quad (6)$$

According to detailed balance principle

$$R_{ei}=\frac{N_{WL}}{N_{Di}}\exp[-E_{Ai}/(KT)]C_i. \quad (7)$$

N_{WL} is effective density of state, N_{Di} is the QD density of ith QD families, E_{Ai} is QD activation energy.

In the model, R_i is endowed with the same value R. With eight parameters g, C, R, R_e', R', E_A, N_{WL}, and N_D existing in Eq. (6) which are substituted with its own value, we can obtain the PL intensity curve of bimodal QD arrays in different QD families depending on temperature. For example, in InAs/GaAs QDs, we simulate the varying relationship of the PL intensity in very dense QD arrays depending on temperature, and we choose the parameter value referring to Ref. [5] which is shown in Table 1.

The effect that QD arrays influence the PL intensity depending on temperature is discussed further, and the curve of PL intensity depending on temperature in low density QD arrays is simulated. It is well known that the change in arrays density will change the capture rate of QD. Therefore, according to the QD arrays of different density, we should change the value of QD capture rate according to Ref. [8] and is shown in Table 2.

3 Result and discussion

The energy band in bimodal QD arrays is shown in Fig. 2.

Large size QD families correspond to the lower ground state energy level. The activation energy E_{A1} between ground state and wetting layer is larger, and the smaller ones compared with the smaller E_{A2}.

Choosing the parameter of QD1 and QD2 in Table 1, we simulate the temperature characteristics of PL intensity in high density bimodal QD arrays, and the result is shown in Fig. 3.

From Fig. 3, we can see that the temperature characteristics of PL in small size QD2 is similar to that of single QD, but the result is different in large size QD1. As temperature increases, the PL is thermal-quenching in small size QD2, and after that, as temperature increases further, the PL intensity has remarkable enhancing tendency in large size QD1, which is thermal-quenching.

Analyzing the curve simulated above, the dynamic process of bimodal QD arrays is obtained in different temperatures: when the temperature is relatively low (0–50 K), carriers are confined inside the QDs, and the number of

Table 1 Parameters used in theoretical simulation of dense QD arrays

quantum	capture rate	combination rate	non-radiation rate	dot density	generation rate	state density of wetting layer	thermal excitation rate
i	C/Hz	R/Hz	R'/Hz	N_D/cm^{-2}	g/Hz	N_{WL}/cm^{-2}	R_e'/Hz
QD1	1×10^{13}	1×10^{9}	1×10^{11}	1.7×10^{11}	1×10^{5}	4.7×10^{17}	1×10^{14}
QD2	0.8×10^{13}	1×10^{9}	1×10^{11}	3×10^{10}	1×10^{5}	4.7×10^{17}	1×10^{14}

Table 2 Parameters used in theoretical simulation of low density QD arrays

N_{D1}/cm^{-2}	N_{D2}/cm^{-2}	C_1/Hz	C_2/Hz
2×10^7	3×10^6	1×10^7	0.8×10^7

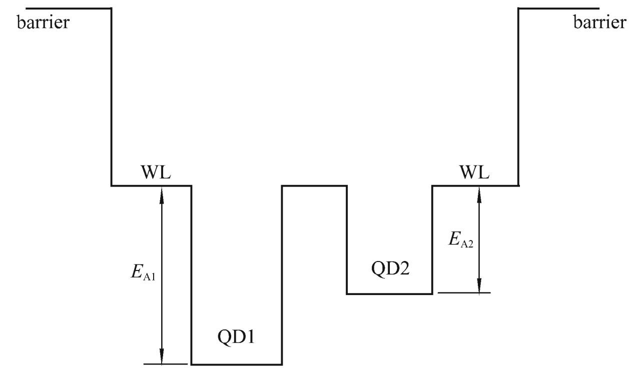

Fig. 2 Schematic band diagram of bimodal QD arrays

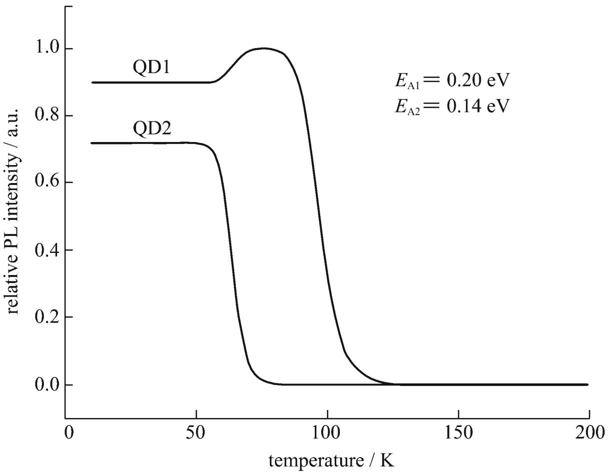

Fig. 3 Temperature dependence of PL intensity of dense QD arrays

carriers stays constant; when the temperature increases to a certain value, the probability of carriers in the small QD families thermally excited to wetting layer is bigger. The number of carriers will decrease with the thermal excitation power, so the intensity of PL decays exponentially. At the same time, the carriers excited into the wetting layer are recaptured by the large size QD1 at a lower energy level, and then the number in the QD1 increases so that the intensity accretes to some extent. That is, as the temperature increases, the carriers will transfer from small QD to large QD, which results in redistribution in different QD families. As the temperature increases further, carriers in QD1 will be excited into the wetting layer, and the PL intensity can decay and finally quench.

Zhang Y C et al. [9] made use of the molecular beam epitaxy technique to grow the InAs/GaAs QDs under the similar condition shown in Ref. [5], and the pattern and PL spectra were researched. The size of QD was a bimodal distribution. The PL spectra in the range from 15 K to 300 K proved that the PL spectral of bimodal QDs presents a bimodal distribution with double Gauss-peak, which corresponds to QDs of different sizes. The temperature dependence of PL intensity presents a similarity to the simulated result theoretically. The intensity of higher energy peak (corresponding to small size QD) decays exponentially as temperature increases, while intensity of lower energy peak (corresponding to large size QD) keeps its shape in the low temperature, after temperature increases to 50 K, which begins to accrete to some extent, decays and finally quenches, which is consistent with our simulated result theoretically.

Choose the capture rate parameter of QD1 and QD2 as in Table 2, the else parameters stay the same as in Table 1. We simulate the temperature dependence of PL intensity in low density QD arrays, and the result is shown in Fig. 4.

Different from the temperature dependence of PL intensity of high density bimodal QD arrays, the PL intensity of large QD1 is totally similar to that of single QD [10]. PL intensity does not show accretion as temperature increases in high density QD arrays. This proves that the process that the carriers transfer in different QDs through the wetting layer in low density QD arrays does not exist. It is considered that the transport of the carriers is limited because of the potential barrier of the wetting layer and interface stress [11]. As the density of QD decreases, coupling between the different families decays gradually until it disappears, which is consistent with the result of Dai Z H [12]. Based on the above analysis, the theoretical model is also suitable in low density QD arrays.

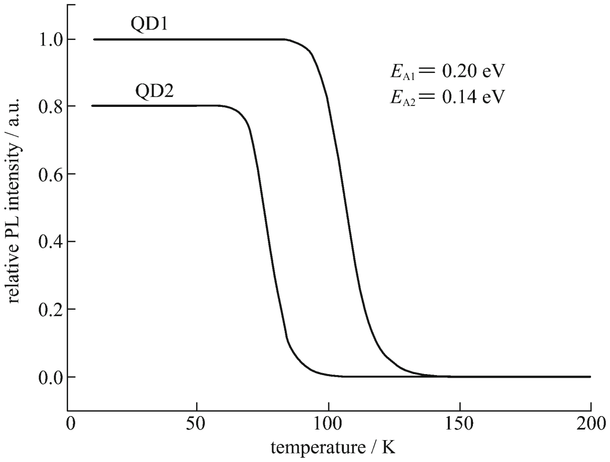

Fig. 4 Temperature dependence of PL intensity of low density QD arrays

Based on the above result, we further study the temperature characteristics of the ratio of the PL intensity in QD arrays and discuss the effect that activation has on the temperature dependence. In fact, the change of activation

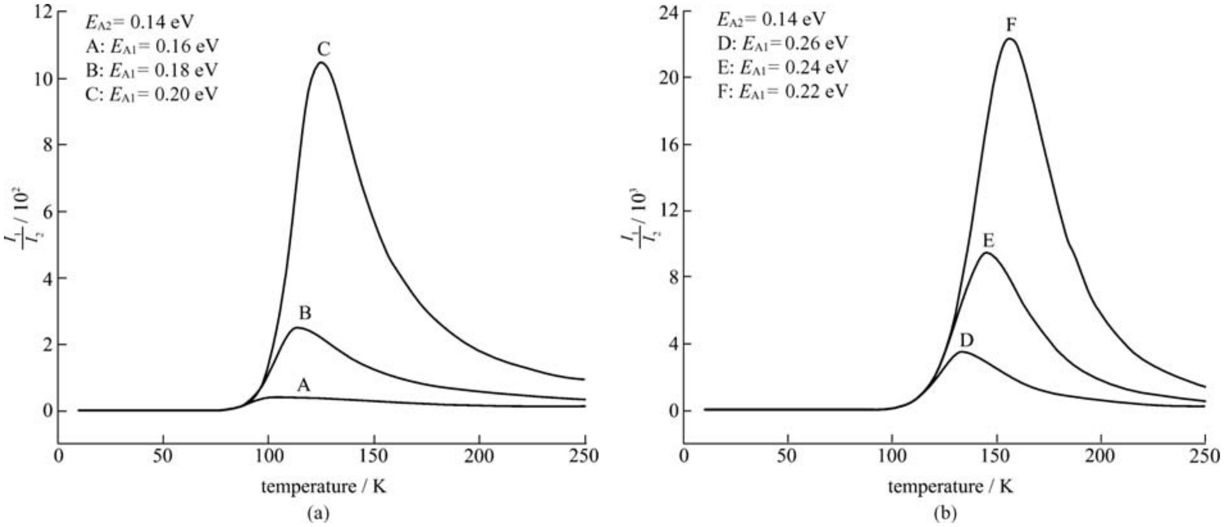

Fig. 5 Temperature dependence of the ratio of PL intensity of bimodal QD arrays, when fixing E_{A2} and adjusting E_{A1}

energy presents the change of the QD size. In the high density QD arrays system, supposing that the activation energy of small QD $E_{A2} = 0.14$ eV is fixed [5], when we adjust the activation of large QD E_{A1}, that is, change the difference of the activation energy of the two kinds of QD, the temperature dependence relationship of the ratio of the PL intensity I_1/I_2 in bimodal QD arrays can be obtained (shown in Fig. 5), and the parameters chosen are in Table 1.

From Fig. 5, as the activation energy of large QD accretes constantly, the ratios of the PL intensity between the QD families increases rapidly as the temperature increases. In multimodal QD, the variation of ratios depending on temperature reflects the distribution of carriers in different size QDs.

The dependence relationship of the maximum ratio of the PL intensity in two QD families on the difference of activation energy ΔE_A can be generalized. Figure 6 shows the relationship of maximum ratio of PL intensity in two QD families and ΔE_A, with the ΔE_A increasing, maximum ratio of PL intensity increases exponentially. It proves that the probability of carriers transferring from the small QD to large QD increases as the activation energy of two families enlarges.

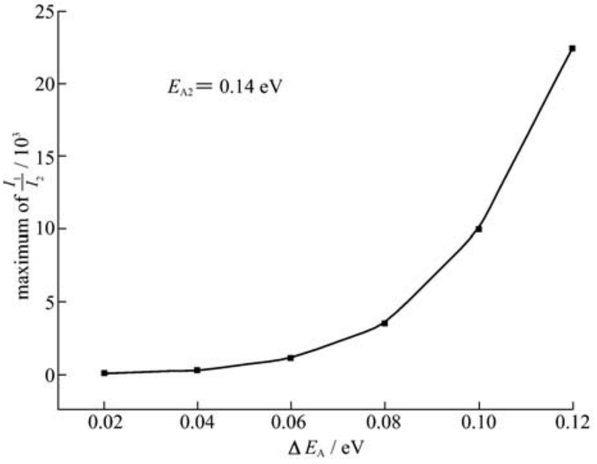

Fig. 6 Maximum of PL intensity ratio as a function of ΔE_A

4 Conclusion

This paper adopts the rate equation model to simulate the thermal quenching of PL in bimodal QD arrays and dependence relationship of the ratios of PL intensity in different QD families. The result shows that in high density bimodal QD arrays, there is a process where the carriers transfer from large QD of high energy to small QD of low energy through the wetting layer, while in low density QD systems, the transfer of carriers between the QD is limited to a great extent. The ratios of PL intensity of QD families depend heavily on the activation energy difference of QD in different temperatures.

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