Fermi approach to the dimension effect of multiple exciton generation in nanostructures

M.B. Marasulov[*,1], N.N. Turaeva[2], B.L. Oksengendler[1,3]

[1]Institute of Polymer Chemistry and Physics, Uzbekistan Academy of Sciences, A. Kadiry str. 7-b, 100128, Tashkent, Uzbekistan
[2]Webster University, USA470 E. Lockwood Ave. St. Louis, MO 63119, USA
[3]Arifov Institute of Ion-plasma and Laser Technologies of Uzbekistan Academy of Sciences. Durmon yuli st. 33, 100125, Tashkent, Uzbekistan

Received 02.06.2021

* Corresponding author: e-mail: polymer@academy.uz, Phone: +998(71)241-85-94

Эффект множественной генерации экситонов в низкоразмерных наноструктурах одним фотоном представляет собой возможный путь к повышению эффективности фотоэлектрического преобразования. В этой работе статистическая теория Ферми генерации сильно коррелированных частиц используется для объяснения размерного эффекта генерации множественных экситонов в наностержнях PbSe по сравнению с квантовыми точками. Статистический подход, учитывающий разницу в пороговых энергиях фотонов для размножения носителей в наностержнях по сравнению с квантовыми точками, дает хорошее согласие с экспериментальными значениями квантовых эффективностей.

Ключевые слова: размерность, множественная генерация экситонов, PbSe, квантовые точки, наностержни, теория Ферми.

The effect of multiple exciton generation in low dimensional nanostructures by a single photon presents a possible route to improved photovoltaic conversion efficiency. In this work, the Fermi`s statistical theory of the generation of strongly correlated particles is used to explain the dimensional effect of the generation of multiple excitons in PbSe nanorods versus quantum dots. The statistical approach taking into account the difference in threshold photon energies for carrier multiplication of nanorods versus quantum dots gives a good agreement with the experimental values of quantum efficiencies.

Keywords: dimension, multiple exciton generation, PbSe, quantum dots, nanorods, Fermi theory.

DOI: https//doi.org/10.52304/.v23i4.286

I. Introduction

One of the remarkable nanoeffects occurring in semiconducting nanocrystals is the generation of multiple electron-hole pairs (excitons) upon the absorption of a single photon [1-8]. The effect called Multiple Exciton Generation (MEG) or Carrier Multiplication (CM) is potentially promising to increase the efficiency of solar energy conversion, particularly in photovoltaics and photocatalysis. The Shockley-Queisser (SQ) detailed balance analysis limits the power conversion efficiency to 33% for a single light-absorbing layer. However, the generation of multiple excitons upon the absorption of a single photon could potentially increase the power conversion efficiency to above 40% [3, 7]. The effect was first observed in bulk semiconductors [9] when a hot electron in the conduction band collides and forces a valence electron to promote across the bandgap, but its probability is very low due to the fast relaxation and phonon emissions. In semiconducting nanocrystals, due to the quantum confinement of electrons and holes, phonon emissions are suppressed, and conservation of translational momentum is relaxed. As a re-

sult, the threshold photon energy E_{th} at which the quantum yield (QY) for exciton generation begins to exceed unity, and the energy of creation of an electron-hole pair $\varepsilon_{e\text{-}h}$ which is the excess photon energy required to generate an additional electron-hole pair after the threshold photon energy, are reduced, and the quantum efficiency of the MEG effect is enhanced.

The first experimental observation of MEG was reported in different quantum dots, including PbSe [1], PbS [2, 6], PbTe [10], Si [5], CdSe [4], InAs [11, 12]. Many theoretical investigations [13-19] followed the experiments to explain the MEG effect. The QYs measured initially by different research groups in identical materials were contradictory [8, 20-23]. These discrepancies were explained by the uncontrolled photocharging of nanocrystals due to the longer lifetime of the photoionized state compared to the time between laser pulses. After eliminating photocharging effects, it was established a good agreement between data obtained by different spectroscopic techniques [20, 21].

Usually, the QYs measurements [24] can be plotted versus $h\nu/E_g$, where $h\nu$ is the photon energy, E_g is the bandgap energy, and the efficiency of an electron-hole pair multiplication $\eta_{e\text{-}h}$ is then obtained from the following relationship between the QY and $h\nu/E_g$ for $h\nu>E_{th}$:

$$\mathrm{QY} = \left(\frac{h\nu}{E_g} - 1 \right) \eta_{e-h} . \qquad (1)$$

At the limit of the efficiency of exciton generation equal to 1, the amount of excess energy needed to produce an additional electron-hole pair is the bandgap energy E_g. For this case, the plot QY vs $h\nu/E_g$ have a "staircase" shape. In reality, the amount of energy for an electron-hole pair creation is greater than the bandgap energy because some excess energy is lost as heat. Since $\eta_{e\text{-}h}$ is defined as the bandgap energy divided by the actual energy of electron-hole pair creation $\eta_{e\text{-}h} = E_g/\varepsilon_{e\text{-}h}$, $\eta_{e\text{-}h}$ is less than 1.

We can compare the values of the efficiency of an electron-hole pair generation for bulk and QDs, for example, for bulk PbSe $\eta_{e\text{-}h}$ = 0.2 while for PbSe QDs $\eta_{e\text{-}h}$ = 0.4 or for bulk Si $\eta_{e\text{-}h}$ = 0.4 and for Si QDs $\eta_{e\text{-}h}$ = 0.9 [24]. In nearly all cases, the efficiencies for the QDs are higher than those for bulk semiconductors. Note here, that the efficiency of exciton generation of (6,5) SWCNTs is equal to 0.7, which is also very promising [25]. In general, two important factors of the MEG effect, the threshold photon energy and the energy of an electron-hole pair creation, are both reduced in QDs compared to those in bulk counterparts and responsible for the enhanced MEG effect in QDs.

The enhancement of the MEG effect due to the reduction of the energy of an electron-hole pair creation is especially promising to obtain an appreciable improvement in the power conversion efficiency of practical devices. As it was shown [24], the smaller energy of an electron-hole pair creation corresponding to stronger MEG can be achieved either by slowing the intra-band energy loss or enhancing Coulomb interactions responsible for exciton-generating collisions.

The dimension of nanostructures is chosen as one of the factors to boost further the MEG performance in PbS and PbSe nanoparticles. According to CM measurements [26-27], multiexciton yield shows an approximately two-fold increase from ~11% in PbSe QDs to ~25% for nanorods for the aspect ratio of ~7. It was shown that at similar threshold photon energy, nanorods exhibited a two-fold reduction of the energy of an electron-hole pair creation over QDs attributed to increased Coulombic coupling in the quasi-1D structure at the optimal aspect ratio of 6-7 [26-27]. At those optimal aspect ratios, the energy of an electron-hole pair creation $\varepsilon_{e\text{-}h}$ in PbSe nanorods is 2.7 E_g and the CM threshold E_{th} is 2.6 E_g, while in PbSe QDs they are 4 E_g and 2.7 E_g [26-27], respectively. It was supposed that decreasing the quantum efficiency at larger aspect ratios is likely due to the restored 1D translational momentum conservation which imposes additional restrictions on the CM process [26-27].

II. Multiple Exciton Generation models

A few models interpret the CM effect in QDs [15-19]. In the impact ionization model [15] the CM was considered inverse with respect to Auger recombination of biexciton states into a single exciton state. Therefore, two time constants should be comparable, or the CM should proceed even slower than that of the Auger decay. However, the transient absorption (TA) data shows that the characteristic CM time is much shorter (<50–200 fs) than the Auger recombination time (~300 ps) indicating that CM is probably not an Auger type process.

According to the coherent CM model [16], due to the superposition of a single-exciton and coherent

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multi-exciton states, coherent oscillations between various resonant-exciton and multi-exciton states should be established. These oscillations were not experimentally detected, instead, the 1S-state build-up is dominated by the intraband relaxation process.

Schaller and et al. [17] showed, that due to the instantaneous character (~200 fs) of generation of multiexcitons they interpreted the MEG in terms of their direct photogeneration of multiexcitons by a single photon via virtual single exciton states. Second-order perturbation theory was applied to the generation of biexciton by a single photon via a virtual single-exciton state by using the electron-electron Coulomb interaction coupling matrix. While coherent CM occurs in the regime of strong exciton-to-biexciton Coulomb coupling, the perturbation approach is valid for the situation of weak to moderate coupling. Although the perturbation theory was applied for the generation of biexcitons, for the processes of high multiplicity it could be problematic [27] due to the strong correlation of multiple excitonic states in the confined space. From another side, it was well-known [28, 29] that for the processes of multiple generation of strongly correlated particles statistical approaches give a more realistic description. The statistical theory developed by Fermi [29-31] was originally developed for the generation of π-mesons at nucleon-nucleon collisions. Due to the large values of interaction constant of the nucleons with π-meson field, for the analysis of the process of generation of π-mesons, the perturbation theory was not justified to be used [28]. Instead, it was supposed that the energy released at nucleon-nucleon collision was instantaneously distributed according to the statistical laws between various degrees of freedom available in the given effective volume. This approach was assumed to be an extreme case inverse to the perturbation theory and a good approximation of the real picture, especially at high energies. In our previous works [18, 19], we used the Fermi statistical approach to the MEG effect in QDs, by assuming the energy of generation of an electron-hole pair to be equal to the energy of semiconductor bandgap (E_g), which is an ideal case of generation of particles without energy losses. With such an assumption, almost ideal high multiplicities were obtained in PbSe QDs [19], which did not correspond to the data on CM observed in the recent experiments [24-27]. In this work, the statistical approach was modified using the real threshold photon energy and additional electron-hole pair creation energy calculated from the experiments [26, 27] to explain the dimensionality effect of CM efficiencies. We compare the quantum efficiencies of MEG in PbSe nanorods versus quantum dots calculated within the Fermi approach with the corresponding experimental results [24-27].

III. Dimension effect in the Fermi approach

By applying Fermi`s statistical theory [29] to the MEG effect, we can suppose, that a high-energetic photon $h\nu >> E_g$ is absorbed in the volume of nanoparticles and is rapidly distributed among the various degrees of freedom present in the system, so that n electrons and holes ($n/2$ excitons) are generated. By neglecting the conservation of momentum, the expression for the probability of generation of n particles with a certain energy distribution in the effective volume Ω can be written as

$$S(n) = \frac{m_0^{3n/2}\Omega^n}{2^{3n/2}\pi^{3n/2}\hbar^{3n}} \frac{T^{(3n/2)-1}}{((3n/2)-1)!}. \qquad (2)$$

Here, m_0 is the free electron mass; $n = 2, 4, 6, ...$ is the number of particles (electrons and holes); T is the total kinetic energy of generated particles. The relative probability of generation of n particles is then determined as

$$W(n) = \frac{S(n)}{\Sigma_n S(n)}. \qquad (3)$$

The QY of CM upon absorption of a single photon is calculated as $QY = \langle N_{e-h} \rangle \times 100\%$, where $\langle N_{e-h} \rangle$ is the average number of electron-hole pairs calculated via statistical weights

$$\overline{n} = 2\langle N_{e-h} \rangle = \frac{\Sigma_n n S(n)}{\Sigma_n S(n)}. \qquad (4)$$

In general, according to the model (Eq. 2), two parameters, the effective volume and the kinetic energy of the particles, vary with the size and shape of nanoparticles. Therefore, the dimension effect observed in experiments [26, 27] can be interpreted based on different values of the effective volume of nanorods versus QDs and the kinetic energy of the particles generated in that volume. The effective volume of QDs is calculated using $\Omega = (3/4)\pi R^3$ and for nanorods it is defined from $\Omega = \pi R^2 L_{eff}$, in which L_{eff} is the effective length of the nanorod, which for PbSe is taken at optimal aspect ratios of 6-7, at which the CM efficiency was maximal [26].

As it was mentioned above, a threshold photon energy, above which QYs begin to exceed 1, and energy of electron-hole pair creation in nanorods is smaller than those in QDs, so in the nanorods, the ki-

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netic energy of the particles should be greater. Thus, the higher efficiency of MEG in PbSe nanorods established from the experiments [27] over QDs at similar bandgap energy can be explained due to the higher values of the effective volume in which multiple particles are created and their total higher kinetic energy. Estimations of QYs for PbSe nanorods and QDs can be made using Eqs. (2, 4). The statistical weight of generation of a single exciton is calculated by using the following formula

$$S(2) = \frac{m_0^3 \Omega^2}{8\pi^3 \hbar^6} \frac{T^2}{5!}. \quad (5)$$

The statistical weight of generation of biexciton is then equal to

$$S(4) = \frac{m_0^6 \Omega^4}{64\hbar^{12}} \frac{T_1^5}{5!}. \quad (6)$$

The kinetic energy of a single exciton generated upon excitation by a high energetic photon (Fig. 1) is calculated from the formula

$$T = h\nu - E_g + E_{exc}. \quad (7)$$

Here E_{exc} is the exciton potential energy. When the photon energy is higher the threshold, an additional exciton is generated, and the kinetic energy of two excitons (Fig. 1) can be calculated based on the following formula

$$T_1 = T - E_{th} + E_{exc}. \quad (8)$$

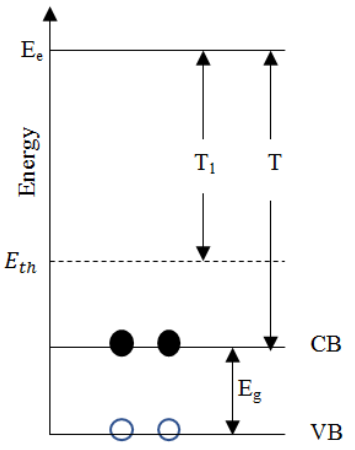


Figure 1. The kinetic energies of a single exciton (T) and biexciton (T_1) are determined via the excitation electron energy (E_e), the bandgap energy (E_g), and the threshold photon energy (E_{th}) The exciton binding energy is not shown.

IV. Results

CM efficiencies in PbSe nanorods and QDs with similar diameter and effective bandgap were calculated using Eq. 4. We compared the theoretical results for nanorods and QDs of three different diameters (2.7 nm, 3.7 nm, and 4.7 nm) with the corresponding experimental ones at 3.1 eV excitation. It was shown (Table 1 and 2) that a good agreement of theoretical results of CM efficiencies for the two types of nanoparticles (d=2.7 nm, and 3.7 nm) with the corresponding experimental data was obtained using the CM threshold photon energy of E_{th}=2.7E_g for QDs, and E_{th}=2.6E_g for nanorods established in the experiments [26, 27]. For the largest nanoparticles (d=4.7 nm) (Table 3), the agreement with the experiments was achieved using E_{th}=3.1E_g. In the calculations of effective volumes of nanorods, the optimal aspect ratio of nanorods (AS=7) was used, at which the CM efficiency was maximal. Based on the dielectric formalism [32], the Coulomb interaction in an electron-hole pair can be approximately defined by the sum of polarizing waves with different wavenumbers, and therefore we assume that the dielectric constant in QDs and nanorods should be smaller than that in bulk semiconductors. Therefore, we chose the arbitrary values for Coulomb interaction energy in the electron-hole pair in PbSe nanorods and QDs which match the model with the experiments.

Note that its value in the nanoparticles is larger than that in bulk PbSe. Moreover, it can be seen from Tables 1-3, the chosen exciton energies inversely correlate with the diameter of the particles.

Table 1. PbSe QDs and nanorods with 2.7 nm in diameter and 0.89 eV bandgap energy.

n	$S(n)$	$\langle N_{e\text{-}h}\rangle_{theor}$	$\langle N_{e\text{-}h}\rangle_{exp}$ [27]
QDs: E_{th}=2.7 E_g; E_{exc}=0.4 eV			
2	2.06×10^{22}	1.16	1.15
4	4.02×10^{21}		
Nanorods: E_{th}=2.6 E_g; L=7×2.7 nm=18.9 nm; E_{exc}=0.4 eV			
2	1.66×10^{24}	1.38	1.30
4	9.84×10^{23}		

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Table 2. PbSe QDs and nanorods with 3.7 nm in diameter and 0.81 eV bandgap energy.

n	$S(n)$	$\langle N_{e\text{-}h}\rangle_{theor}$	$\langle N_{e\text{-}h}\rangle_{exp}$ [27]
QDs: E_{th}=2.7 E_g; E_{exc}=0.17 eV			
2	1.21×10^{23}	1.23	1.22
4	3.65×10^{22}		
Nanorods: E_{th}=2.6 E_g; L=7×3.7 nm=25.9 nm; E_{exc}=0.02 eV			
2	1.18×10^{25}	1.55	1.5
4	1.47×10^{25}		

Table 3. PbSe QDs and nanorods with 4.7 nm in diameter and 0.66 eV bandgap energy.

n	$S(n)$	$\langle N_{e\text{-}h}\rangle_{theor}$	$\langle N_{e\text{-}h}\rangle_{exp}$ [27]
QDs: E_{th}=3.1 E_g; E_{exc}~0 eV			
2	5.0×10^{23}	1.4	1.50
4	3.58×10^{23}		
Nanorods: E_{th}=2.6 E_g; L=7×4.7 nm=32.9 nm; E_{exc}=0 eV			
2	5.52×10^{25}	1.89	1.80
4	4.34×10^{26}		

V. Conclusion

The statistical model is presented to explain the dimensionality effect on CM established in the experiments [26, 27]. According to the model, the larger kinetic energy due to the smaller energy threshold of CM results in higher quantum yields of MEG in nanorods compared to those in QDs which is in good agreement with the experiments. The model takes into account the enhancement of exciton energy in low dimensional nanostructures. It can be used to estimate the CM efficiency in 0D and 1D nanostructures based on the photon energy threshold and energy of the creation of additional electron-hole pairs above the energy threshold.

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Наноструктураларда кўп экситон генерациясининг ўлчам эффектига Ферми ёндашуви

М.Б. Марасулов[1], Н.Н. Тураева[2], Б.Л. Оксенгендлер[1,3]

[1]Ўзбекистон Республикаси Фанлар Академияси Полимерлар кимёси ва физикаси институти, А.Қодири кўчаси 7б, 100128, Тошкент, Ўзбекистон
[2]Вебстер университети, USA470 E. Локвуд Ави. Ст. Лоуис, МО 63119, АҚШ
[3]Ўзбекистон Республикаси Фанлар Академияси Ионплазма ва лазер технология институти, Дўрмон йули, 33, 100125, Тошкент, Ўзбекистон

Кичик ўлчамли наноструктураларда битта фотон билан кўп экситон генерацияси эффектини фотоэлектрик ўзгариш самародорлигини оширишнинг мумкин бўлган усулини ўзида намоён қилади. Ушбу ишда PbSe наностержен кўп экситон генерацияси ўлчам эффектини квант нуқталар билан солиштиришда Ферминиг кучли корреляция қилинган заррачалар ҳосил бўлишининг статистик назариясидан фойдаланилади. Наностерженларда квант нуқталар билан солиштирганда ташувчиларни кўпайиши учун фотон чегара энергияси фарқини хисобга олувчи статистик ёндашув квант самарадорлигининг экспериментал қийматлари билан яхши мос келади.

Калит сўзлар: Ўлчовлилик, кўп экситон генерацияси, PbSe, квант нуқталар, наностержен, Ферми назарияси

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