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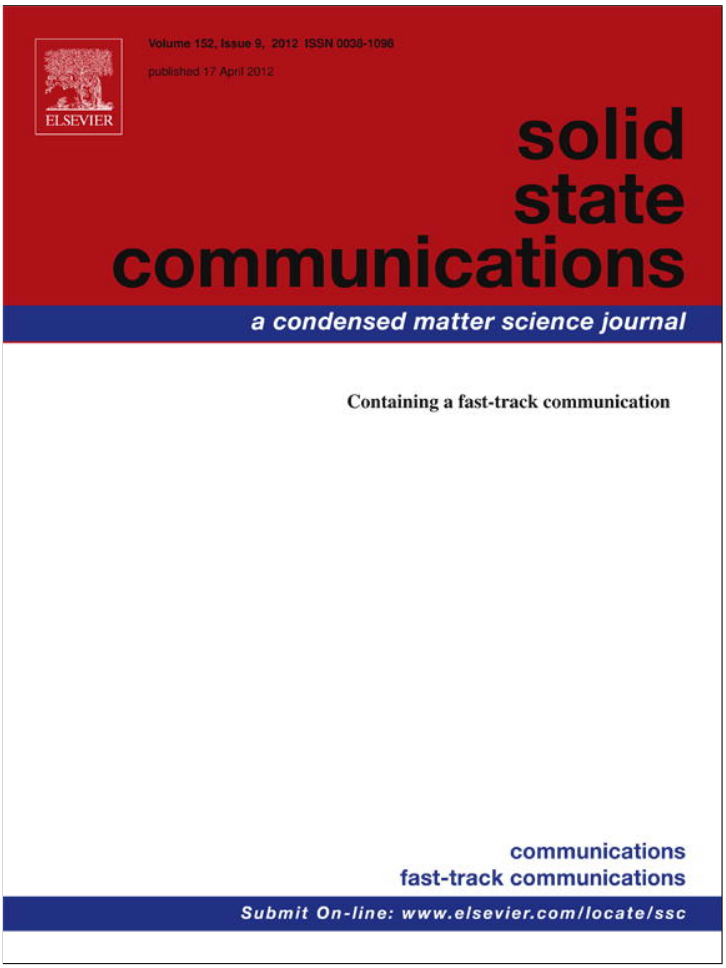

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A statistical understanding of multiple exciton generation in PbSe semiconductor nanostructures

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ABSTRACT

We propose a simple statistical model, based on Fermi statistical theory and impact ionization mechanism, to resolve the controversies over the effects of multiple exciton generation (MEG) in PbSe quantum dots (QDs). We have confirmed that MEG indeed occurs in PbSe QDs. Also, we have found out that there exists a critical radius R_c (∼9 nm) such that the MEG efficiency of PbSe QDs is smaller than that of the bulk counterpart if $R < R_c$, but larger if $R > R_c$. Moreover, we have found out that the MEG threshold energy calculated for PbSe QDs shows a universal behavior. The present work provides a powerful theoretical means not only for further experimental investigations into the MEG effects in semiconductor nanostructures, but for their applications in photovoltaic devices.

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Multiple exciton generation (MEG), the creation of multiple electron–hole pairs per single photon absorption, is not only of fundamental importance [1], but of practical importance in quantum dot lasers [2] and highly efficient photovoltaics [3–5]. Although MEG in bulk semiconductors has been well understood in terms of impact ionization [1] since 1950s, it has not been thought to be practical for applications due to the very low MEG efficiency [6,7]. However, since highly efficient MEG in PbSe semiconductor quantum dots (QDs) has been reported in 2004 [8], MEG in semiconductor nanostructures has quickly become a very attractive research subject. A great number of experimental studies have been followed to show such highly efficient MEG in semiconductor nanocrystals or QDs such as PbSe [8–14], PbS [10,12], PbTe [15], CdSe [11], Si [16], and InAs [17]. Also, MEG has been demonstrated to play an important role in quantum dot photodetectors [18] and carbon nanotube photodiodes [19], implying that the MEG effects may be useful for applications in highly efficient photovoltaic devices. In fact, quantum yields have recently been reported to be greater than one electron per photon via MEG in a sensitized photovoltaic system [20].

Despite MEG has become very important in optoelectronic applications, there still remain unsolved controversies over experimental/theoretical results of the MEG effects in semiconductor QDs. These controversies can be summarized as follows: (i) does MEG occur in semiconductor QDs? i.e., some claimed it does [8–19,21,22], whereas others did not [23,24]; if it does, (ii) does the MEG efficiency of QDs is larger or smaller than that of bulk materials [8,12,25]? and (iii) is there any explanation for differently observed results on the MEG threshold energy in the same QDs [8,11,12,14,15,26–28]? Some experimental [9,21] and theoretical (virtual exciton generation approach [27], coherent multiexciton model [10,29], and atomistic model [30–32]) works have been elaborated to clarify the controversies. Nevertheless, none of those efforts has yet resolved them, completely. In this work, we propose a simple statistical model, developed within the framework of Fermi statistical theory [33] and impact ionization [1], to explore MEG in semiconductor nanostructures, especially in PbSe QDs. With our model, we have been able to well explain the MEG effects observed in PbSe QDs, such as the size-dependent MEG efficiency and threshold energy.

In semiconductor QDs of the band gap E_g, the absorption of a single photon of high-energy $h\nu > E_g$ (h is the Planck constant and ν photon frequency) may generate an electron–hole pair (or an exciton). In the case of $h\nu > 2E_g$, the nascent photogenerated charge carriers may generate the second exciton via collision with other bound electrons, which is so-called impact ionization. If the nascent charge carriers after collision and/or collision-generated

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charge carriers have enough kinetic energy, they may generate more excitons again via the collision, leading to the creation of multiple excitons [1,30,34]. In this impact ionization process, the incident photon energy is released into the energy relaxation volume Ω (a volume within which excitons are generated) of QDs for a ultra-short time amount t_s (50–200 fs) [27], leading to a statistical equilibrium final state with n particles (n is even; $n/2$ electrons and $n/2$ holes, or $n/2$ excitons). Note that, in semiconductor QDs, Auger recombination can be ignored in this impact ionization process due to its long recombination timescale (∼hundreds of picoseconds).

Although there are Coulomb interactions between electrons and/or holes generated, these charged particles can be treated as quasi-independent particles in semiconductor materials including large QDs. This quasi-particle treatment is valid, since the generation of one particle does not affect the positions and the momenta of other generated particles, i.e., if n particles are generated in a quantum dot by absorbing a photon, we cannot determine the position and the momentum of a particle through determining the positions and the momenta of the other $n-1$ particles. This quasi-particle treatment is still valid even in small QDs ($R < R_B$, R_B is the Bohr radius of exciton), in which, even though Coulomb interactions will be enhanced, the quantum confinement becomes so strong that Coulomb interactions can be neglected. In this strong confinement approximation, electrons and holes can be treated as free particles [35,36]. In fact, this quasi-particle treatment has been widely employed in low-dimensional semiconductor systems such as QDs [36].

In QDs, by using the effective mass approximation, the kinetic energy E_k can be written as $E_k = m^*_{n,p}v^2/2$ in terms of the electron (hole) velocity v, where $E_k \le h\nu - E_g$, $m^*_{n,p}$ are the effective masses of electron and hole, respectively. And, the largest electron (or hole) energy relaxation volume V_{er} for the energy relaxation time t_s, for which multiple excitons are generated, depends on its longest energy relaxation length $l_\varphi = (l_{fp}vt_s/2)^{1/2}$ that corresponds to the maximum speed of electron (or hole), where l_{fp} is the free path of electron (hole) in QDs and it can be expressed as $V_{er} = \pi l^3_\varphi/6$. Considering the fact that electron (or hole) cannot move outside QDs, the energy relaxation volume Ω, therefore, should be the smaller one among the largest energy relaxation volume V_{er} and the total quantum dot volume V, i.e., $\Omega = \min(V_{er}, V)$.

According to Fermi's golden rule, the transition probability of a particle from a given state to another state is proportional to the square of the corresponding effective matrix element, and also to the density of equilibrium final states per unit energy interval. The square of the effective matrix element is proportional only to a probability $p(n)$ that n particles in the equilibrium state are contained within the energy relaxation volume Ω at the same time. As a result, the statistical weight $\omega(n)$ in QDs can be expressed as the product of $p(n)$ and the density of equilibrium final states $S(n)$. Note that $S(n)$ is the number of equilibrium final states per energy interval in the $n/2$-exciton system, which is different from the electron density of states in QDs.

In a quantum dot of the volume V, if $n/2$ excitons (i.e., n particles) with momenta $\vec{P}_1, \vec{P}_2, \ldots, \vec{P}_n$ are generated by the absorption of a single photon, $p(n)$ and $S(n)$ can be given by Fermi [33]:

$$p(n) = \left(\frac{\Omega}{V}\right)^n; \qquad S(n) = 2 \times \left(\frac{V}{8\pi^3\hbar^3}\right)^n \frac{dQ(E_{ks})}{dE_{ks}}, \tag{1}$$

where the factor 2 comes from the electron spin, $\hbar(= h/2\pi)$ the reduced Planck constant, $Q(E_{ks})$ the volume of the $3n$-dimensional momentum space corresponding to the total kinetic energy of the system E_{ks} ($E_{ks} = P_1^2/2m_1 + P_2^2/2m_2 + \cdots + P_n^2/2m_n$ with m_1, $m_2, \ldots, m_n$ the effective masses of n independent particles), and $dQ(E_{ks}) = dP_{1x}dP_{1y}dP_{1z}\cdots dP_{nx}dP_{ny}dP_{nz}$ the differential element of volume. Then, Eq. (1) can be written as:

$$S(n) = 2 \times \frac{(m_1m_2\cdots m_n)^{3/2}V^n}{2^{3n/2}\pi^{3n/2}\hbar^{3n}} \frac{E_{ks}^{3n/2-1}}{(3n/2-1)!}. \tag{2}$$

Since the total kinetic energy of the system E_{ks} is equal to $(h\nu - nE_g/2)$, resulting from the energy conservation, the statistical weight $\omega(n)$ can then be expressed as:

$$\omega(n) = p(n) \times S(n) = 2 \times \frac{(m^*_n m^*_p)^{3n/4}\Omega^n}{2^{3n/2}\pi^{3n/2}\hbar^{3n}} \times \frac{(h\nu - nE_g/2)^{3n/2-1}}{(3n/2-1)!}, \tag{3}$$

where the effective mass approximation was also taken into account. Note that our statistical weight $\omega(n)$ contains two variable parameters: the energy relaxation time t_s and the band gap E_g.

Due to the quantum size effect, the energy gap E_g is closely related to the total quantum dot volume V that is equal to $4\pi R^3/3$ with the quantum dot radius R in the case of a spherical quantum dot. Hence, the energy gap E_g is given in terms of R by Brus [37,38]:

$$E_g = E_{g0} + \frac{h^2}{8R^2}\left(\frac{1}{m^*_n} + \frac{1}{m^*_p}\right) - \frac{1.78e^2}{\varepsilon R}, \tag{4}$$

where E_{g0} and ε are the band gap and the dielectric constant of bulk materials, respectively, and e is the electron charge. Here, the second and third terms are the (additive) quantum confinement energy and the Coulomb (attraction) energy, respectively.

Now, the maximum number of excitons N is equal to $[h\nu/E_g]$, where the square bracket denotes the integer part of $h\nu/E_g$. Thus, the relative probability $W(n)$ can be calculated by:

$$W(n) = \frac{\omega(n)}{\sum_{n=2,\text{even}}^{2N}\omega(n)} \quad (n = 2, \ldots 2N), \tag{5}$$

and the statistical average number of electron–hole pairs $\langle N_{exc}\rangle$ can then be calculated by:

$$\langle N_{exc}\rangle = \frac{1}{2}\sum_{n=2,\text{even}}^{2N} nW(n). \tag{6}$$

Since the MEG efficiency or inner quantum efficiency I_{QE} in semiconductor materials can be defined as the average number of excitons created by the absorption of a single photon [39], it can be expressed as:

$$I_{QE} = \langle N_{exc}\rangle \times 100\%. \tag{7}$$

Note that the MEG efficiency I_{QE} calculated can be said a *pure* MEG efficiency in a sense that our calculation do not includes multi-photon absorption effects and defect trapping of electrons and holes. If there is no confusion, we shall use the MEG efficiency for this *pure* MEG efficiency, hereafter. Also, since we do not consider the quantum transition selection rules in our calculation, the MEG efficiency calculated is expected to be larger than the experimental value, however, it will turn out that it is close to each other, as will be shown later.

We firstly consider PbSe QDs of $E_g = 0.65$ eV. The radius R of the QDs can be obtained to be 3.90 nm from Eq. (4), in which all the required parameters for calculation were taken from Ref. [40]. For $h\nu = 3.1$ eV and $t_s = 50$ fs, the relative probabilities $W(n)$ are 0.193, 0.778, 2.877×10^{-2}, and 3.559×10^{-7} for 1-, 2-, 3-, and 4-exciton generations, respectively, from Eq. (5). Thus, we obtain from Eq. (6) that the statistical average number of excitons $\langle N_{exc}\rangle = 1.836$. This readily results in that the MEG efficiency $I_{QE} = 183.6\%$

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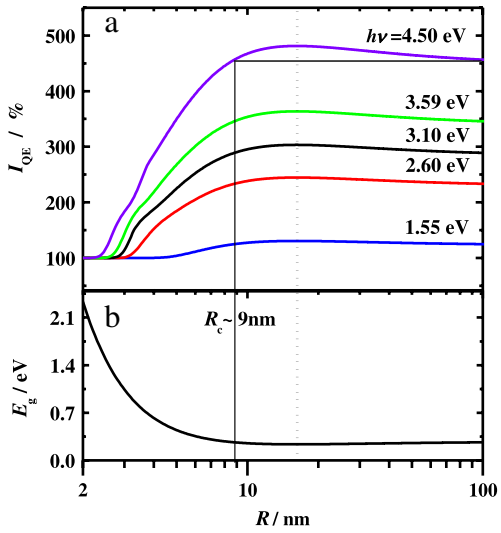

Fig. 1. (a) Calculated I_{QE} versus R for various photon energies at $t_s = 50$ fs in PbSe QDs. (b) Calculated E_g versus R in PbSe QDs. The dotted line indicates the maximum I_{QE} in (a) and the minimum E_g in (b) at $R = 16.20$ nm. The critical radius R_c is also indicated.

from Eq. (7). As for $t_s > 50$ fs, we found that I_{QE} is gradually increased to ~200% with increasing t_s up to 70 fs, and remains almost unchanged with further increasing t_s up to 200 fs. This calculated I_{QE} (183.6%) is in good agreement with an accurately obtained experimental result (170%) in Ref. [21], in which multi-photon absorption and defect trapping have been carefully ruled out, and in which the same band gap and photon energy have been employed as in our calculation. Note that our calculating result is somewhat higher than the experimental value, resulting from the fact that our model has to yield the largest MEG efficiency, as discussed earlier.

Now, let us discuss the controversies over the MEG effects in literature [8–10,12,21–27], as mentioned earlier. In Fig. 1(a), we show I_{QE} as a function of quantum dot radius R for various photon energies at a fixed $t_s = 50$ fs in PbSe QDs. For a given incident light, as can be seen in the figure, I_{QE} is rapidly increased with increasing R and reaches the maximum value at $R = 16.20$ nm (dotted line), and, it is then very slowly decreased with further increasing R up to 100 nm (~5.5 times the Bohr radius of excitons in PbSe QDs), at which these large QDs can be considered as bulk materials. Note that the Bohr radius of excitons in PbSe QDs, R_B, can be estimated to be 18.10 nm by using the effective masses of charge carriers and the dielectric constant of PbSe taken from Ref. [40].

According to Ref. [41], if $R \leq 2R_B = 36.20$ nm, the quantum confinement is so strong that it cannot be neglected, whereas, if $R \geq 4R_B = 72.40$ nm, it becomes so weak that it can be. In QDs, Coulomb attraction has to be considered in addition to the quantum confinement. In fact, there is a competition between the Coulomb energy and the quantum confinement energy, both of which will contribute to the size-dependent band gap, $E_g = E_g(R)$, as shown in Eq. (4). The Coulomb energy clearly tends to become weaker with increasing R. Moreover, even with decreasing R, its enhancing tendency ($\sim R^{-1}$) appears to be dominated by a stronger enhancement of the quantum confinement energy ($\sim R^{-2}$), resulting in Fig. 1(b), in which the R-dependent E_g of PbSe QDs is exhibited. With increasing R, E_g is rapidly decreased to its minimum value (~0.24 eV) again at $R = 16.20$ nm (dotted line), and it is then negligibly increased above $R = 16.20$ nm, reminiscent of a bulk material behavior. Note that this rapid drop of E_g for small QDs results in a significant enhancement of the MEG efficiency.

Here, we want to comment on the most fundamental question, i.e., whether or not does MEG occur in semiconducting QDs? As can be seen in Fig. 1(a), MEG actually occurs in PbSe QDs, since the MEG efficiency is always bigger than 100% for $h\nu > 2E_g$, indicating that there always exist non-zero probabilities of multiple exciton generation.

In some experimental studies on PbSe small QDs [8–10,12,22], in which $R = 2.4$–4.2 nm, the MEG efficiency has been claimed to be much higher in QDs than in bulks, due to the enhanced Coulomb attraction. Note here that, even for those small QDs, the enhancement of Coulomb attraction may not play an important role, as discussed above. On the other hand, others studies, including a tight binding calculation ($R = 2.5$–3.7 nm) [31] and a recent experimental observation ($R \sim 1$ μm, i.e., bulk) [25], have presented that the MEG efficiency in QDs is close to or a little smaller than in bulks. However, as can be seen in Fig. 1(a), the MEG efficiency I_{QE} that is in fact a function of the dot radius R can be described as follows: it is smaller in QDs than in bulks if $R < R_c$, but slightly-yet-clearly larger if $R > R_c$, where the critical radius R_c comes from an assumption that PbSe dots of $R = 100$ nm are of bulk behavior. As indicated in the figure, $R_c \sim 9$ nm in PbSe, remaining almost unchanged even at a much bigger R, due to a very small change in I_{QE} for a large R (>100 nm). It is worth pointing out that this critical radius may play an important role in designing high-efficient QDs that can be exploited to develop ultra-efficient photovoltaic devices.

Returning to Fig. 1(a), in addition to the size effect of I_{QE}, the figure also shows that, for a given quantum dot, i.e., at a fixed R, the larger the photon energy, the higher the MEG efficiency. This can be readily understood in terms of three following aspects of the photon energy effect. First, the larger photon energy may lead to the larger energy relaxation volume. Second, the larger photon energy may lead to the larger maximum number of excitons. Third, the large photon energy may lead to the enhancement of the density of equilibrium final states. In fact, these three independent aspects of the photon energy effect are reflected in Eq. (7) to calculate the MEG efficiency, readily leading to the fact that the higher MEG efficiency results from the larger photon energy for a given quantum dot.

Addressing the controversy over the MEG threshold energy E_T (defined as the lowest photon energy required to make MEG to occur, independent of the incident photon frequency) in literature, particularly for PbSe QDs, it has been reported to be ~2.8 eV [8], 5.1 eV [11], 1.8 eV [12], and so on, as shown in Fig. 2(a). Also, a band structure calculation [26] has predicted that the normalized MEG threshold energy by the band gap, E_T/E_g, depends on E_g, whereas a numerous number of experimental studies [8,11,12,14,15,27,28] have claimed that it is nearly independent of E_g. It has also been claimed by some groups [8,10] that the normalized energy E_T/E_g has to be a constant for the same material. However, different results have been obtained such as ~3.0 [8] and ~2.1 [10].

In Fig. 2(a), we present our calculated E_T (solid curve) in PbSe QDs at a fixed $t_s = 50$ fs, and we also present the results reported in literature (squares). Our calculation shows that E_T significantly depends on R. With increasing R, it is rapidly decreased down to the minimum value (~0.95 eV) again at $R = 16.20$ nm, and it is then negligibly increased, similar to the behavior of the R-dependent E_g in Fig. 1(b). This similarity can be readily understood from the fact that the wider band gap needs the higher photon energy for MEG. Most importantly in the present work, almost all the experimental results available in literature [8,11,12,14,15,27,28] are excellently consistent with our calculated E_T, showing a universal behavior of E_T. This also indicates that E_T certainly depends on R, i.e., E_T shows a strong dependence of E_g, consistent with the band structure calculation [26] as mentioned above.

Finally, we show in Fig. 2(b) E_T/E_g as a function of E_g, showing that E_T/E_g is not a constant even though it is nearly a constant (~2.3) above a characteristic band gap E_{gc} (~1.04 eV). This scaled

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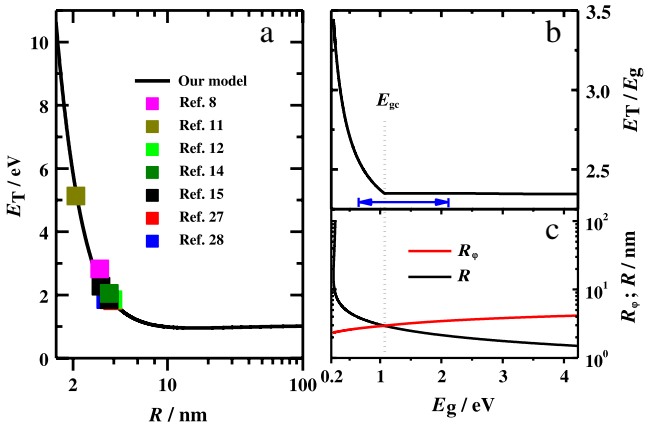

Fig. 2. (a) Calculated E_T versus R at $t_s = 50$ fs in PbSe QDs (solid line). The solid squares are experimental data in literature. (b) E_T/E_g versus E_g in PbSe QDs. (c) The largest energy relaxation radius R_φ ($= l_\varphi/2$) numerically calculated versus E_g at $t_s = 50$ fs for the photon energy $h\nu = E_T$. $R = R(E_g)$ from Eq. (4) is also plotted. The dotted line and the double arrow indicate the characteristic band gap E_{gc} and the investigated E_g range of experiments in literature, respectively.

MEG threshold energy strongly depends on E_g for a large R, i.e., for $E_g < E_{gc}$, whereas it is nearly independent of E_g for a small R, i.e., for $E_g > E_{gc}$. This nearly independent behavior for a small size quantum dot appears to explain why the E_g-independent E_T/E_g has been obtained in the above-mentioned experimental studies [8,11,12,14,15,27,28], whose investigated range of E_g is indicated by a double arrow in Fig. 2(b). The calculated E_g-dependent E_T/E_g relies on the fact that the statistical weight of 2-exciton, $\omega(2)$, has to be a finite constant for a given quantum dot, and, at the same time, it results from a consideration in our model that the energy relaxation volume Ω is the smaller one among V_{er} and V, i.e., $\Omega = \min(V_{er}, V)$. In Fig. 2(c), we show the largest energy relaxation radius $R_\varphi (= l_\varphi/2, l_\varphi =$ the longest energy relaxation length) as a function of E_g for the incident photon energy $h\nu = E_T$ at $t_s = 50$ fs, and also show the E_g-dependent R obtained from Eq. (4). Now, $\omega(2) \sim \Omega^2 E_g^2 (E_T/E_g - 1)^2$ for $h\nu = E_T$ from Eq. (3), which has to be a finite constant. If $E_g < E_{gc}$, for which $R_\varphi < R$ as in Fig. 2(c), then $\Omega = V_{er}$ that is decreased as E_g is decreased, obviously resulting in a rapid increase of E_T/E_g with decreasing E_g. In contrast, if $E_g > E_{gc}$, for which $R < R_\varphi$ as in the figure, then $\Omega = V$ that is decreased as E_g is increased, leading to a nearly E_g-independent E_T/E_g for a high E_g due to the compensation between E_g and Ω.

In summary, we have used Fermi statistical theory and impact ionization mechanism to develop a simple statistical model. This simple model has turned out to well explain MEG in semiconductor PbSe material including both quantum dot and bulk. This simple yet powerful model has allowed us to resolve perhaps all the controversies over MEG in PbSe QDs: i.e., MEG indeed occurs, which rules out the existence controversy; there exists a critical radius R_c, below (above) which the MEG efficiency is smaller (larger) in QDs than bulks; and the MEG threshold energy depends on the size of the dots, which universally explains almost all the different results available in literature. Finally, it is worth noting that our simple statistical theory can be applied to describing MEG in any semiconductor nanostructures.

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