Carrier multiplication in bulk and nanocrystalline semiconductors: Mechanism, efficiency, and interest for solar cells

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Carrier multiplication (CM), the possibility to generate more than one exciton in a semiconductor quantum dot (QD) after absorption of a single photon has been intensely debated in recent years. Following on previous theoretical and experimental work, we report here that: (1) although the CM factor (i.e., number of generated photons per absorbed photon) at a given photon energy is higher in bulk than in QDs of the same material [Pijpers *et al.*, Nature Phys. **5**, 811 (2009)], the energy efficiency (the relative fraction of the photon energy that is transformed into excitons rather than heat) is higher in QDs; (2) for the same ~1.2 eV band gap, CM is more efficient in PbSe QDs than in bulk silicon; (3) nonetheless, the efficiency of solar cells based on PbSe QDs is not significantly enhanced by CM compared to a bulk silicon-based device.

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I. INTRODUCTION

Semiconductor quantum dots (QDs) are considered promising building blocks for new generations of solar cells because their optical properties can readily be tailored by changing their composition, size, and shape. The interest in QDs was further sparked by observations of efficient carrier multiplication (CM),[1] i.e., the generation of two or more excitons after absorption of a single photon. In bulk semiconductors, CM relies on a high-energy photon that produces several electron-hole pairs or excitons upon relaxation of the photoexcited carriers by impact ionization. This process is rather inefficient in bulk semiconductors and it was argued that CM is enhanced in QDs[2] and indeed, after the first report by Schaller *et al.*,[1] several experimental studies confirmed the occurrence of efficient CM in PbSe,[1,3–6] PbS,[3] CdSe,[6,7] InAs,[8,9] and Si (Ref. 10) QDs.

Very high CM factors (number of excitons per absorbed photon) have been previously reported (up to 7)[4] and therefore theoretical models were proposed to explain these intriguing results.[4,11] But the existence of high CM factors in QDs has been recently questioned both theoretically and experimentally. Theoretical efforts have shown that the relaxation of excited carriers by impact ionization may explain modest CM factors in QDs[12–16] but cannot explain the highest CM factors reported experimentally,[12,13] even in the extreme situation where carriers cannot relax by emission of phonons.[12] On the experimental side, Nair *et al.*[17] have found little to no evidence for CM in CdSe and CdTe QDs in contrast to previous works.[6,7] In the most recent studies on PbSe (Refs. 18–21) and InAs (Refs. 8 and 22) QDs, the measured CM factors are appreciably lower than in early reports, and therefore it was no longer evident that QDs exhibit higher CM factors than their bulk counterpart. Very recently, measurements on bulk PbSe and PbS (Ref. 23) have demonstrated that the CM factor at a given photon energy, is in fact larger in bulk than in corresponding QDs.

The present work continues on the results presented in Refs. 12, 13, and 23. New here is: (i) an explanation of the methodology used to calculate the CM factor in bulk PbSe and large PbSe QDs; (ii) the demonstration that the thus calculated CM factors are in agreement with the majority of the reported experimental values for bulk PbSe and PbSe QDs (for all investigated QD sizes); (iii) a detailed discussion on the *absolute* CM factor at a given photon energy vs the *energy efficiency* of the process. This discussion is closely related to the discussion regarding the use of absolute or relative photon energy scales: in literature, the CM factor has been plotted on either an absolute or a relative energy scale. We argue here that the use of an absolute energy scale is more appropriate when considering the physics underlying the CM process, but that the relative energy scale is more insightful when discussing the implications of CM in the context of possible device performance. In this context, we define the quantity *energy efficiency* as the ratio between the total excitonic energy (the average number of generated excitons times the energy gap) and the photon energy $h\nu$. The energy efficiency is the relevant parameter when determining the interest of CM in QDs for, for example, photovoltaic devices, since the energy efficiency corresponds to the relative fraction of the photon energy that is transformed into excitons instead of heat after relaxation of hot carriers. We demonstrate that the CM factor of PbSe QDs with a 1.2 eV energy gap (ε_g) is larger than the CM factor in bulk silicon (ε_g also ~1.2 eV), clearly indicating that the *energy efficiency* is higher in nanostructured semiconductors (at a given energy gap). However, we also show that CM only marginally improves the efficiency of QD solar cells, despite the higher-energy efficiency of PbSe QDs with a 1.2 eV band gap compared to bulk silicon.

Summarizing, this paper presents an integrated picture of the CM process from QDs to bulk semiconductors: we show that the current data for both bulk and QDs can be fully accounted for by CM theory based on impact ionization. The

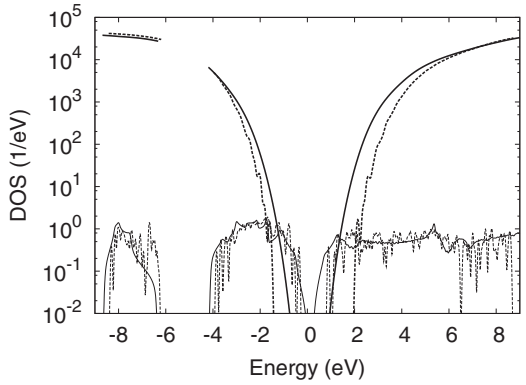

FIG. 1. DOS per atom versus the energy of the excited carrier for bulk PbSe (solid lines) and PbSe QDs with an energy gap of 1.2 eV (dashed lines). The thin lines represent the DOS of initial states and the thick lines correspond to the DOS of final states. The zero of energy corresponds to the top of the bulk valence band.

analysis reveals that variations in density of states (DOS) can account for both the higher absolute CM factor in bulk and the higher energy efficiency in QDs. We present a discussion of the implications of these observations for photovoltaic applications.

II. METHODOLOGY

The methodology of the calculations follows that reported in our previous work on the role of impact ionization in the CM process in small PbSe QDs (Refs. 12 and 13) and in bulk PbSe.[23] Here, we demonstrate how this treatment is extended to QDs of arbitrary size. In the following we present a brief overview of the theoretical analysis.

The absorption of a high-energy photon in a QD results in the generation of hot carriers (either electrons or holes). We model the relaxation of these hot carriers to the ground state as a competition between two relaxation processes, i.e., relaxation via impact ionization and relaxation via sequential emission of phonons.[12,13,23] The relative rates of these competing processes determine the final CM factor, which is defined as the number of generated electron-hole pairs (excitons) per absorbed photon. We calculate the electronic structure of the QDs in tight binding and study the relaxation of electrons and holes by impact ionization. As can be seen in Fig. 1, the density of initial states to which carriers can be excited is relatively independent of the energy, but the density of final states increases strongly with energy of hot carrier. For electrons (holes) excited in the conduction (valence) band, the rate of relaxation by impact ionization was calculated using Fermi's golden rule. For higher electron energies, the impact ionization rate increases steeply because of the increased density of final states (see Fig. 1),[12,14,15] leading to an increase in the number of relaxation pathways via which impact ionization can proceed. We have observed previously[12] that the impact ionization rate, when summed over all possible final states after relaxation, strongly depends on the initial energy of the carrier and vanishes when the carrier excess energy (the energy of the carrier with respect to the band edge) is equal to, or less than, the QD gap. Another important result of previous studies[12] is that, at high excess energy, the magnitude of the impact ionization rate and its dependence on excess energy are almost the same in QDs and in bulk. However, at low excess energy (i.e., close to the QD gap energy), the impact ionization rate is reduced in QDs with respect to the bulk material because of the smaller density of final states.[12,13] This effect is clearly visible in Fig. 1, where the density of final states at relatively low energies is orders of magnitude lower in QDs than in bulk material. The simulations include all possible initial energies of photogenerated carriers and all possible impact ionization relaxation channels, and provide the number of excitons generated after absorption of a photon when impact ionization competes with (sequential phonon) intraband relaxation.

This treatment closely mimics the true relaxation process of hot carriers but is computationally very demanding, due to the large number of possible channels for the relaxation of a carrier by impact ionization. In our previous works,[12,13] the calculation of the impact ionization rate could be performed for small QDs and bulk semiconductors. For bulk, a limited set of states was used by sampling the Brillouin zone, but for QDs, all the final states were considered. This procedure is only possible for small QDs because the number of final states is limited: it increases as the third power of the number of atoms.[13] The calculation of the CM factor was restricted to small QDs (for PbSe, diameter below 3.1 nm)[12,13] and could not be directly applied to larger QDs and bulk semiconductors. We have therefore developed a simplified method to extend the calculation of the CM factor to larger QDs and bulk. First of all, the energy-dependent impact ionization rate for larger QDs is obtained by interpolating numerically between the results for small QDs and bulk.[12,13] We then define an energy grid with discrete levels spaced by 5 meV. Note that this energy grid does not represent the actual DOS, but allows us to calculate the CM factor in larger QDs, given a certain energy-dependent impact ionization rate and phonon relaxation rate. The CM factor is calculated assuming that all possible final states (defined by the grid and allowed by energy conservation) can be reached with the same probability, which is given by the total impact ionization rate at the energy of the carrier divided by the number of channels. Concerning the phonon-assisted relaxation, we consider that each carrier can decay to lower energy by emission of a phonon of energy $\hbar\omega_{\mathrm{ph}}$ with a rate $1/\tau_{\mathrm{ph}}$. For reasons discussed in Ref. 12, the lifetime τ_{ph} is considered as a parameter independent of the energy. In this procedure, the calculation time is independent of the system size.

The interpolation procedure to obtain the impact ionization rates for large QDs and the approximation of the QD electronic structure by a 5 meV grid are justified because the influence of the final DOS is implicitly included in the variation of the total impact ionization rate with energy and the CM factor does not depend on the details of the initial DOS. The results of this new procedure are in excellent agreement with our previous fully explicit simulations for PbSe and Si QDs.[12,13]

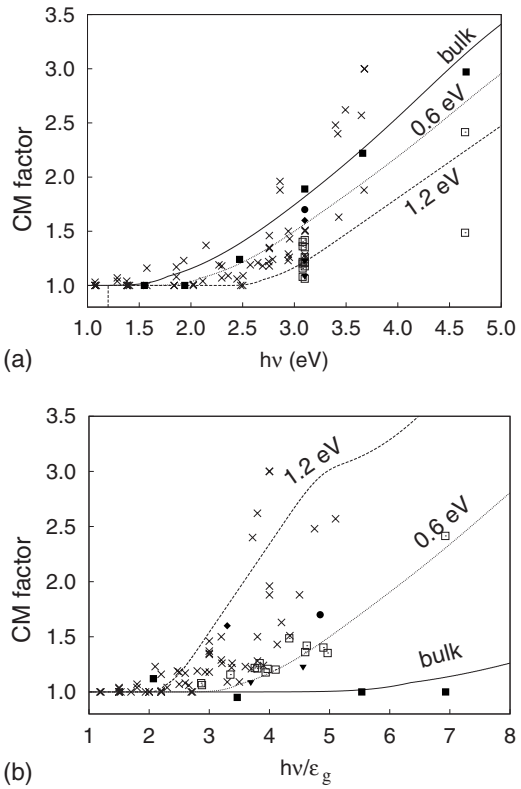

FIG. 2. CM factor: Number of excitons generated by impact ionization after the absorption of a photon of energy $h\nu$. (a) Lines: simulations for bulk PbSe (gap ε_g=0.28 eV) and for QDs (ε_g =0.6 eV, 6.9 nm diameter and ε_g=1.2 eV, 3.1 nm diameter). All the calculations were performed with τ_{ph}=0.5 ps. The symbols indicate the recent experimental results of Ref. 23 (■) for bulk PbSe and those of Ref. 18 (●), Ref. 20 (◆), Ref. 19 (□), Ref. 21 (▼), and Ref. 3 (×) for PbSe QDs. (b) Same data but plotted versus the energy-gap-normalized photon energy, $h\nu/\varepsilon_g$.

III. RESULTS AND DISCUSSION

A. CM factor

Figure 2 presents the results of the simulations for bulk PbSe (gap of 0.28 eV) and two PbSe QDs (gaps of 0.6 and 1.2 eV) on an absolute *and* a relative photon energy scale. For the sake of comparison, all the simulations were performed assuming the same intraband relaxation lifetime τ_{ph} =0.5 ps. With this value, the theory is not only in good agreement with the experimental data of Ref. 23 for bulk PbSe (■, Fig. 2) but also in reasonable agreement with the most recent measurements in QDs (other symbols in Fig. 2). This implies that impact ionization provides a consistent and sufficient explanation as the origin for CM in both bulk and QDs. An impact ionization mechanism was not capable of explaining the higher CM factors reported earlier,[12,13] but there has been some debate concerning the experimental conditions of these early measurements.[17,19,23]

When plotting the CM factor vs an absolute photon energy scale [Fig. 2(a)], the calculations reveal that, when the gap of the system increases, the number of generated excitons decreases and the threshold for the CM is shifted to higher energy. These results are intuitive: the impact ionization rate becomes smaller when the QD size decreases because the density of final states decreases, in particular at low carrier excess energy.[12,13,21] This DOS argument explains why the number of generated excitons is larger in bulk than in QDs,[23] in contrast to previous expectations.[2]

Plotting the number of generated excitons vs the absolute photon energy does not take into account the variations in the band gap occurring for QDs. However, when considering the physics underlying the CM process for PbSe QDs, i.e., if one is interested in the mechanism of CM and the resulting number of electron-hole pairs, there are two reasons to justify this representation on an absolute energy scale. First, given the bulk mechanism of CM as a competition between phonon emission and impact ionization, one would expect the initial excess energy of the carrier (and the associated density of initial and final states) to be the key parameter determining the CM factor, and not how many times this energy fits the band gap. Second, and closely related, it is evident that for the relevant photon energies >3 eV where significant CM occurs, the variation in excess energy of generated hot carriers for QDs of varying sizes is relatively small for a given photon energy, and the CM factor is therefore not limited by limitations due to energy conservation, but rather by variation in the DOS. Such a conclusion would not be apparent from inspection of Fig. 2(b), which shows the same data on a band-gap-normalized photon energy scale. A similar argumentation for using an absolute energy scale has previously been put forward by Nair and Bawendi.[17,21]

The results in Fig. 2(a) demonstrate that, for a given photon energy, the resulting number of carriers is larger in bulk than in QDs. However, the energy of the excitons in QDs is higher than that of the electron-hole pairs in the bulk material. Plotting the exciton yield as a function of band-gap-normalized photon energy [Fig. 2(b)], shows that although the absolute number of generated electron-hole pairs is larger in bulk, the photon energy is used more efficiently for the quantum dots. In contrast to bulk material, the CM factor is larger than 1 at low values of $h\nu/\varepsilon_g$ for QDs. Hence, Fig. 2(b) illustrates clearly that the energy efficiency is higher in QDs than for bulk.

The comparison between theory and experiments in Fig. 2(a) is not fully appropriate because the experimental results have been obtained for different sizes of QDs showing a large distribution of energy gaps. For this reason, we compare in Fig. 3 the results obtained only at the fixed excitation energy $h\nu$=3.1 eV, for which many results are available. Only recent experimental data are presented and we plot the CM factor as a function of the gap. Despite significant variations in the experimental results (which presumably may be traced to differences in QD surface chemistry)[19,24,25] it can be concluded that (1) CM experiments are compatible with (impact-ionization based) theoretical results calculated for τ_{ph} varying between 0.1 and 1 ps, i.e., in a reasonable range and (2) there is a tendency toward a decrease in the CM yield with increasing gap, in agreement with theory.

The agreement between theory and experiments is consistent with the notion that, in QDs, the intraband relaxation by emission of phonons does not slow down significantly due to the phonon bottleneck.[26–29] This latter result is not surprising[21] because: (1) we consider here carriers at high

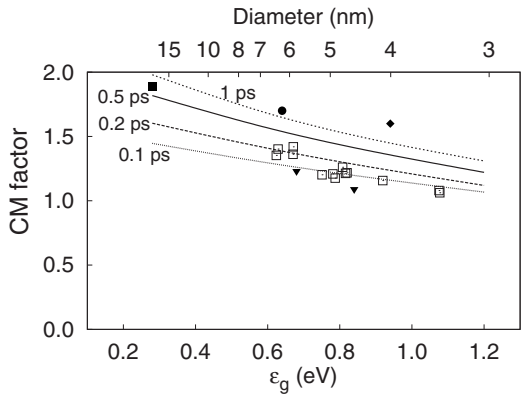

FIG. 3. CM factor: Number of excitons generated by impact ionization in PbSe QDs and bulk PbSe after the absorption of a photon of energy $h\nu$=3.1 eV represented as a function of the energy gap ε_g of the system (ε_g=0.28 eV for bulk PbSe). Lines: simulations for different values of the intraband relaxation lifetime τ_{ph} (0.1, 0.2, 0.5, and 1 ps). The symbols indicate the recent experimental results of Ref. 23 (■) for bulk PbSe and those of Ref. 18 (●), Ref. 19 (□), Ref. 20 (◆), and Ref. 21 (▼) for PbSe QDs.

excess energy in regions characterized by a high DOS; (2) even at low excess energy, the effect of the phonon bottleneck is only visible under very specific conditions (single excited carrier, thick passivating shell, fast electron-hole separation)[28–30] which have not been fulfilled in experiments on CM.

B. Energy efficiency

Since the number of generated excitons is smaller in QDs than in the corresponding bulk semiconductor at a given photon energy, one might be tempted to conclude that there is no motivation to use QDs in solar cells for their CM abilities. However, for photovoltaic applications, the CM factor is not the quantity of interest because the energy of the excitons is not the same in QDs and in the bulk. Rather, a more relevant quantity is the energy efficiency defined as the ratio between the total excitonic energy (the number of excitons times the energy gap) and the photon energy $h\nu$. As mentioned above, the energy efficiency corresponds to the relative amount of energy which is transformed into excitons instead of heat after relaxation of the carriers. In Fig. 4, we plot the energy efficiency (by definition valued between 0 and 1) including (solid lines) and omitting (dashed lines) impact ionization effects. Interestingly, the energy efficiency above the gap is much larger in small QDs than in the bulk. For PbSe QDs with a gap of 1.2 eV, we predict for τ_{ph}=0.5 ps that the energy efficiency is always larger than 50%. Also, the contribution of CM to the energy efficiency is largest for the smallest QDs. Even if the number of generated excitons is smaller in QDs than in the bulk, this is more than compensated by the increase of the excitonic energy due to the larger gap in QDs.

Therefore, strongly confined QDs are energetically relatively efficient for photons with energy above their gap. This conclusion is further confirmed by comparing the CM factor in bulk silicon with PbSe QDs of approximately the same

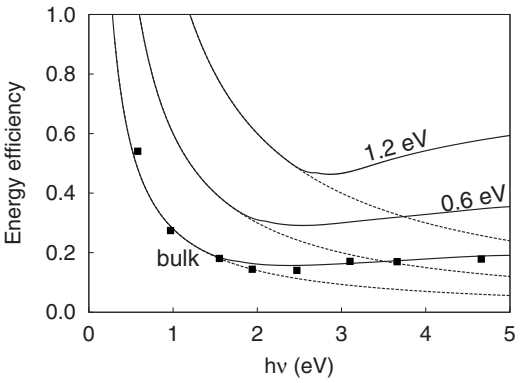

FIG. 4. Energy efficiency of the CM versus photon energy $h\nu$. Thick solid lines: simulations (τ_{ph}=0.5 ps) for bulk PbSe (ε_g=0.28 eV) and for two QDs (ε_g=0.6 eV and ε_g=1.2 eV). Thin dashed lines: same but assuming that there is no impact ionization, i.e., when excited carriers can only relax by emission of phonons. Black squares: experimental results of Ref. 23 for bulk PbSe.

gap (ε_g=1.2 eV). Figure 5 shows that the absolute CM factor of PbSe QDs is clearly superior, the thresholds for impact ionization being more than 1 eV lower in energy. We obtained similar results when comparing bulk silicon with PbS QDs. It is likely a general conclusion that QDs made of a small band-gap semiconductor exhibit higher CM factors than a bulk semiconductor with the same gap as the QDs. The general reason is that the DOS increases similarly with (photon) energy for different semiconductor materials, but from differently positioned starting points, i.e., the conduction and valence-band edge. As reported in Ref. 12 and as shown in Fig. 1, the density of final states for impact ionization is similar for QDs and bulk of the same material for photon energies above the energy conservation threshold. For a given carrier energy, the DOS is larger in bulk PbSe than in bulk silicon, in particular because of the eightfold degeneracy of the valence- and conduction-band extrema.[32] At sufficiently high carrier energy, the DOS for PbSe QDs approaches that of bulk PbSe (Fig. 1) and hence the DOS at

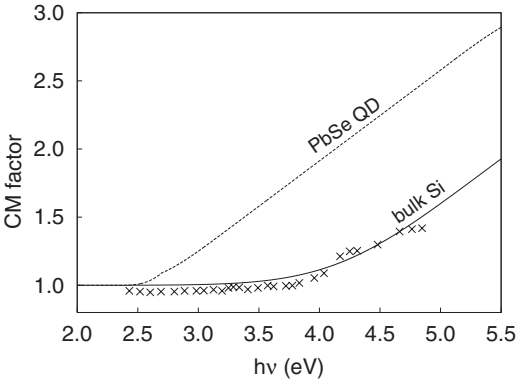

FIG. 5. CM factor: Number of excitons generated by impact ionization after the absorption of a photon of energy $h\nu$. Solid line: simulations for bulk Si (gap=1.2 eV). Crosses: experiments of Ref. 31. Dashed line: simulations for a PbSe QD characterized by a diameter of 3.1 nm and a gap of 1.2 eV. All the calculations are performed with τ_{ph}=0.5 ps.

TABLE I. Energy efficiency calculated (τ_{ph}=0.5 ps) for bulk PbSe (ε_g=0.28 eV) and for two QDs (ε_g=0.6 eV and ε_g =1.2 eV) under solar illumination, in presence or in absence of CM. The last column is the ratio between third and second columns, which represents the average number of excitons generated after absorption of a single photon when the system is irradiated by solar light.

Gap (eV)	Without CM	With CM	Ratio
0.28	0.199	0.217	1.09
0.60	0.381	0.400	1.05
1.20	0.476	0.485	1.02

a given energy is always larger in PbSe QDs than in bulk silicon, despite quantum confinement effects. Furthermore, Fig. 5 shows that also for bulk silicon the calculations are in excellent agreement with the experimental results[31] for a reasonable value τ_{ph}=0.5 ps ($\hbar\omega$=50 meV), once more confirming the validity of the methodology of the calculations.

From the above, it is evident that our theoretical considerations can fully account for the higher energy efficiency in QDs compared to bulk (despite the lower CM factors) by means of DOS arguments.[12] In bulk materials, for energy regions relatively close to the band gap, impact ionization is energetically allowed but has a very low probability. This region of reduced impact ionization is smaller in QDs because of the quantum confinement that opens the gap: for QDs the DOS increases sharply above the band gap toward bulk values (Fig. 1). Above the threshold for impact ionization, the variation in the impact ionization rate is very large in the case of QDs and it quickly reaches the bulk values, because the DOS at high energies increasingly resembles that of the bulk. This explains *both* why the absolute CM factors are lower in QDs compared to bulk, but also why the energy efficiency is enhanced for QDs: as soon as carriers in QDs have an energy slightly above the threshold, they have the same probability to relax by impact ionization as in bulk, but the energy of the additionally generated exciton is larger than in bulk because of opening of the gap due to quantum confinement.

C. Interest of CM for solar cells

Finally, we discuss the implications of our findings on the benefits of CM in QDs for QD solar cells. Despite the observation that PbSe QDs exhibit higher CM factors than bulk silicon, the added value of CM for photovoltaics seems limited. This is demonstrated in Table I, where we calculated the energy efficiency averaged for the solar spectrum

$$\frac{\int_0^{\lambda_g} \lambda/\lambda_g \eta(\lambda) I(\lambda) d\lambda}{\int_0^{\infty} I(\lambda) d\lambda}. \quad (1)$$

$I(\lambda)$ is the wavelength dependent solar intensity for the unconcentrated ASTM G-173-3 reference solar spectrum,[33] $\eta(\lambda)$ is the CM factor and therefore $\lambda/\lambda_g \eta(\lambda)$ is the energy efficiency [$\lambda_g = hc/\varepsilon_g$]. We assume that each photon with energy above the gap is absorbed and that each generated carrier contributes to the photocurrent at the maximum voltage. The absolute increase in photovoltaic efficiency gain induced by CM is at best 9% for small gap devices (bulk PbSe) that are inherently inefficient. For a realistic gap of a photovoltaic device (~1.2 eV), the absolute CM-related gain in the light to current conversion efficiency is limited to 2% when using PbSe QDs. This small value is due to the fact that most of the photon flux in the solar spectrum is below the CM threshold which is, for example, positioned at 2.5 eV for 1.2 eV PbSe QDs (Fig. 5). Therefore, the benefit of CM in QD based photovoltaics is minor, in spite of the relatively high energy efficiency of CM in PbSe QDs.

IV. CONCLUSION

In conclusion, we have shown that recent experimental measurements of CM in PbSe nanocrystals can be fully accounted for by the generation of excitons by impact ionization. Theory and experiments demonstrate that the CM factor (number of generated excitons per absorbed photon) is larger in the bulk semiconductor than in QDs but that the energy efficiency (relative fraction of the photon energy that converted into excitons) is higher in QDs. We have shown that QDs based on small band-gap semiconductors exhibit higher CM factors compared to bulk semiconductors with similar band gap. In spite of that, the increase in efficiency induced by CM is predicted to be small in solar cells based on these QDs. The search for nanoscale systems with improved conversion of solar photons into excitons thus remains an important challenge.

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