Supplementary Information: Optimal quantum dot size for photovoltaics with fusion

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Abstract

Light fusion increases the efficiency of solar cells by converting photons with lower energy than the bandgap into higher energy photons. The solar cell converts the product photons to current. We use Monte Carlo simulation to predict that lead sulfide quantum dot sensitizers will enable fusion with a figure of merit on the $\mathrm{mA\,cm^{-2}}$ scale, exceeding current records, while enabling silicon cell compatibility. Performance is highly sensitive to quantum dot size, on the order of $\mathrm{mA\,cm^{-2}\,nm^{-1}}$.

1 Supplementary Calculations

The behavior of triplet fusion can be summarized by the well-known differential equation:[2]

$$\frac{d[T]}{dt} = k_\phi[S] - k_1[T] - k_2[T]^2 = 0 \tag{1}$$

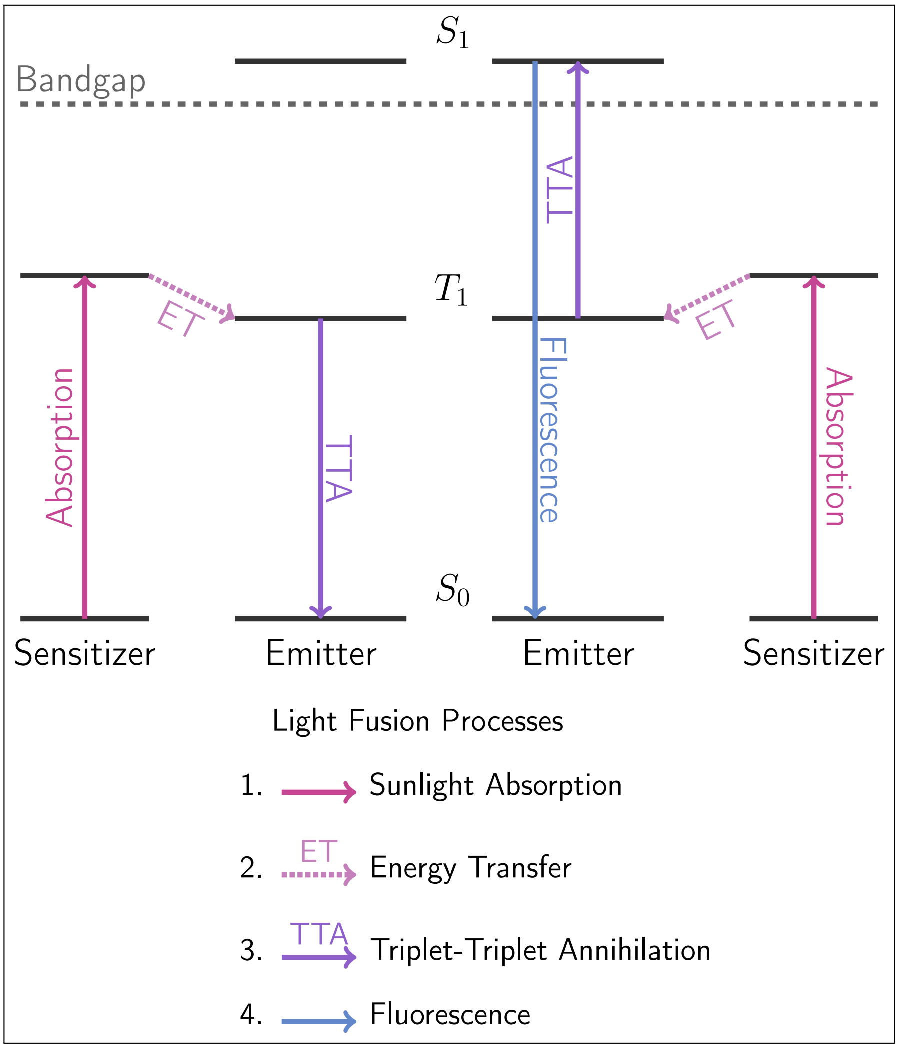

Figure 1: Energy level diagram for light fusion. To be efficient, each process must be exothermic.[1] S_n indicates the emitter molecule's nth singlet state. T_1 indicates the first triplet state.

Where t is time, $[T]$ is the concentration of triplet excitons, k_ϕ is the sensitizer excitation rate, $[S]$ is the sensitizer concentration, k_1 is the decay rate for noninteracting triplet excitons, and k_2 is the annihilation rate constant for triplet excitons. The $k_2[T]^2$ term produces the upconversion. This equation qualitatively explains Figs. 2–5. Figures 2 and 3 show the calculated performance of quantum dot sensitizers when paired with emitters having various properties. Fig. 4 is calculated as a function of illumination conditions. Fig. 5 addresses quantum dot concentration. Collectively, these figures show that a high ($>1\,\mathrm{mA\,cm^{-2}}$) figure of merit can be achieved over a wide range of device types.

Fig. 6 demonstrates that an insufficiently accurate model of the solar spectral irradiance should not be used to inform device design.

In our source for quantum dot molar absorptivity, the quantum dot dispersity is at most 10%.[3] Higher dispersity will reduce the degree to which the figure of merit depends on the quantum dot size. For optimally sized quantum dots, dispersity slightly decreases the figure of merit slightly, as shown in Fig. 7. This is an example of regression towards the mean. We define dispersity as the width of a rectangular distribution of radii centered at 2.2 nm.

In each supplementary figure, the thickness of the device is variable as a function of the horizontal axis. The thickness was selected to produce the highest figure of merit.[4] The quantum dot radius is 2.2 nm. The quantum dot concentration is 0.1 mM, except in Fig. 5.

2 Toxicity

Toxicity is a concern for technologies that may be mass produced. We estimate a lead sulfide density of $6\,\mathrm{g\,m^{-2}}$. The LC_{50} toxicity metric of bulk lead sulfide in the fish *Pimephales promelas* is $0.9\,\mathrm{mg\,L^{-1}}$.[20] We did not locate mammalian toxicity data or any data for PbSe. The ratio of density to LC_{50} is 7 m of water, which indicates that careful device disposal is required. For PbS nanomaterials, surface passivation may substantially reduce toxicity.[21,22]

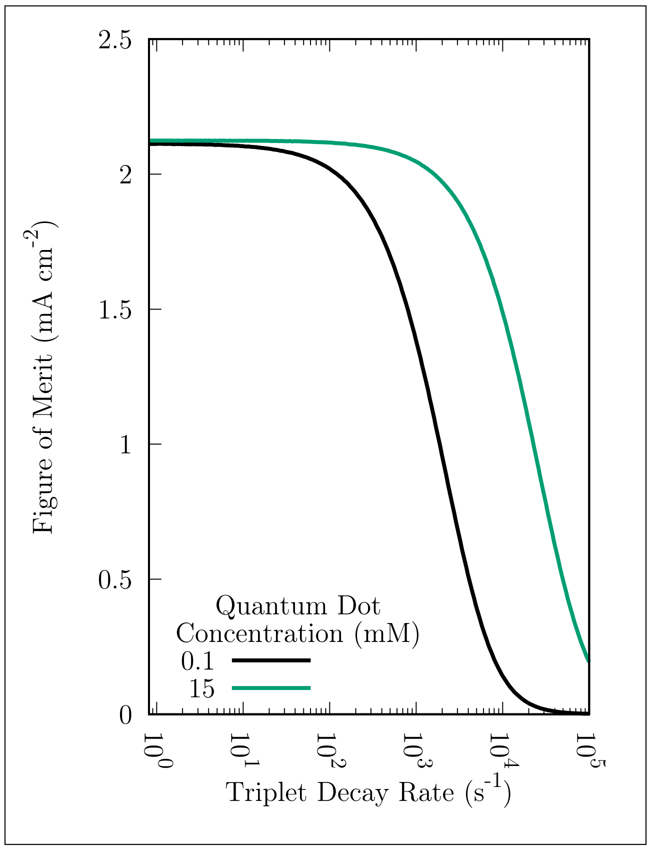

Figure 2: Figure of merit of quantum dot sensitized light fusion as a function of the decay rate for noninteracting triplet excitons. The decay rate is a property of the emitter.[5,6] The decay rate can be as low as $90\,s^{-1}$.[7–9] It can be increased by adding traps[1,7] such as oxygen molecules.

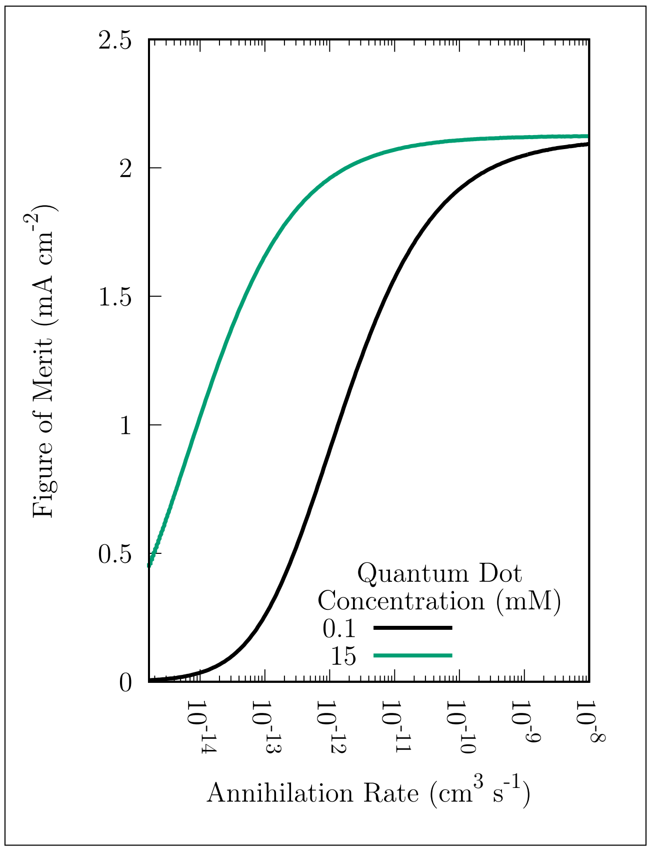

Figure 3: Figure of merit of quantum dot sensitized light fusion as a function of the annihilation rate constant for triplet excitons. The constant is a property of the emitter. The annihilation rate constant varies from $10^{-14}\,\mathrm{cm^3\,s^{-1}}$ to $10^{-9}\,\mathrm{cm^3\,s^{-1}}$.[4,10,11]

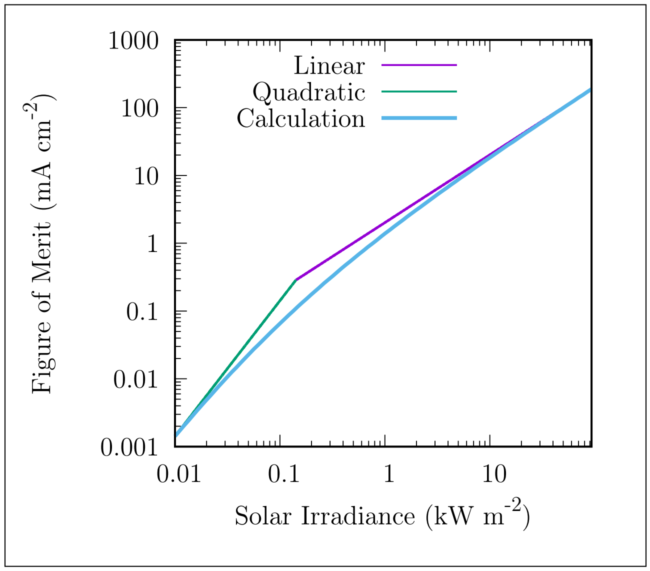

Figure 4: Figure of merit of quantum dot sensitized light fusion as a function of the solar irradiance. The irradiance is measured before the solar cell, not at the sensitizer. $1\,\mathrm{kW\,m^{-2}}$ is the conventional value. The transition from linear to quadratic behavior[12–15] happens near I_{th} =$0.2\,\mathrm{kW\,m^{-2}}$. Quadratic behavior indicates the quantum yield is at a maximum. To the best of our knowledge, the lowest I_{th} reported is $0.09\,\mathrm{kW\,m^{-2}}$,[16] but this is for monochromatic illumination in a silicon-incompatible device.

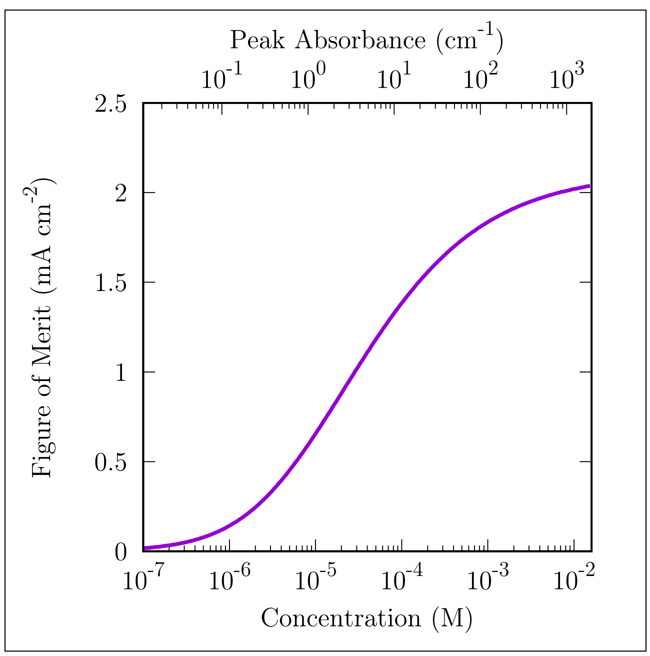

Figure 5: Figure of merit of quantum dot sensitized light fusion as a function of the quantum dot concentration.[17,18] The absorbance of the quantum dot at the lowest energy absorption peak is also indicated. A higher concentration results in a higher figure of merit.

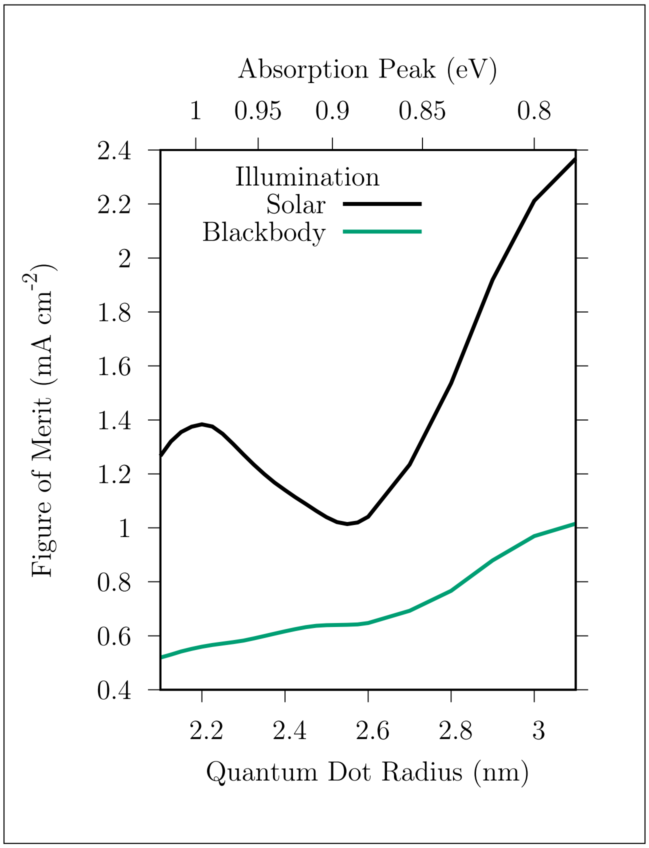

Figure 6: Figure of merit of quantum dot sensitized light fusion as a function of the quantum dot radius. The black curve is the same as main text Fig. 3. The green curve is the same calculation, except the solar spectrum is replaced by a 5778 K blackbody spectrum.[19] The irradiance is held constant at $1\,\mathrm{kW\,cm^{-2}}$. The attenuation by the earth's atmosphere is the main reason the solar spectrum is redder than a blackbody. The blackbody approximation fails to capture the full importance of the quantum dot radius.

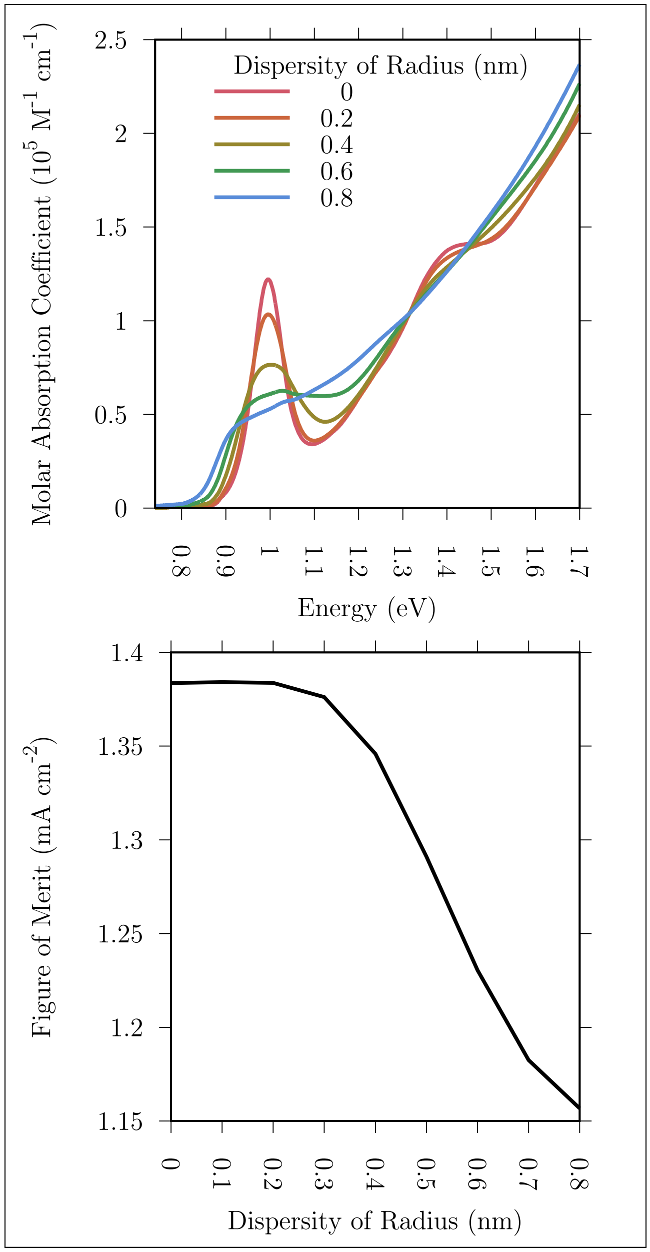

Figure 7: Molar absorption coefficient of quantum dots and figure of merit as a function of the dispersity of the quantum dot radius.

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