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Trap-assisted transport and non-uniform charge distribution in sulphur-rich PbS colloidal quantum dot-based solar cells with selective contacts

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Abstract

This study reports evidences of dispersive transport in planar PbS colloidal quantum dots heterojunction-based devices as well as the effect of incorporating a MoO3 hole selective layer on the charge extraction behavior. Steady state and transient characterization techniques are employed to determine the complex recombination processes involved in such devices. The addition of a selective contact drastically improves the device efficiency up to 3.15 % (especially through the photocurrent and series resistance) and extends the overall charge lifetime by suppressing the main first-order recombination pathway observed in device without MoO3. The lifetime and mobility calculated for our sulphur-rich PbS-based devices are similar to previously reported values in lead-rich quantum dots-based solar cells. Nevertheless, strong Shockley-Read-Hall mechanisms appears to keep restricting charge transport, the equilibrium voltage taking more than 1 ms to be established.

Keywords

contacts, cells, solar, dot, quantum, colloidal, pbs, rich, sulphur, distribution, selective, charge, trap, uniform, non, transport, assisted

Disciplines

Engineering | Physical Sciences and Mathematics

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SUPPORTING INFORMATION

Trap-Assisted Transport and Non-Uniform Charge Distribution in Sulphur-Rich PbS Colloidal Quantum Dot-based Solar Cells with Selective Contacts

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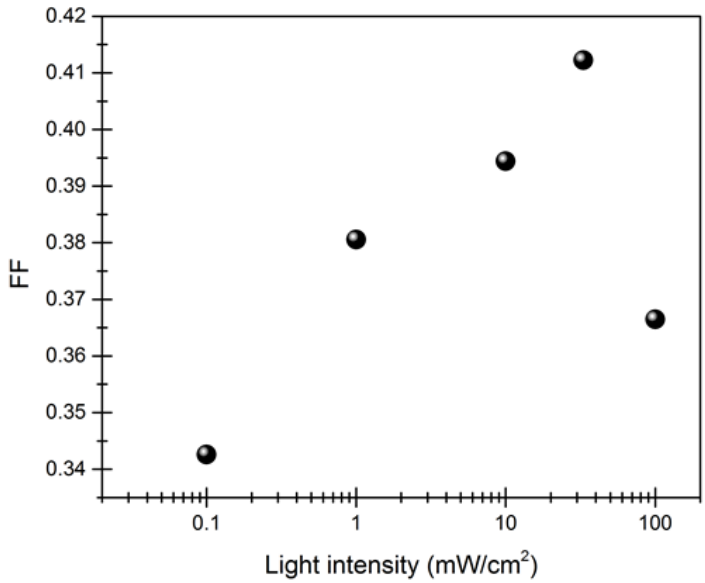

Figure S1. Fill factor (FF) as a function of the light intensity.

Fitting of dispersive exponential

The fitting functions are:

- Geminate: $n(t) = n_0 e^{-\left(\frac{t}{\tau}\right)^{\alpha}}$
- Non-geminate: $n(t) = \frac{n_0}{1+n_0(\beta t)^{\alpha}}$

where n_0 is the initial concentration of free photogenerated charges and τ, β, and α are the geminate recombination characteristic time, the non-geminate recombination coefficient and the dispersive factor. These two functions are obtained from solving the continuity reaction: $\frac{dn}{dt} = \frac{dp}{dt} = -\beta n(t)p(t)$. In the case of geminate recombination, the equation can be simplified as the density of one charge carrier can be considered constant (e.g., $n(t) = n$ and $\beta n = \frac{1}{\tau}$), thus leading to a solution following a monoexponential decay. Typical non-geminate, assuming charge quasi-neutrality ($n(t) \approx p(t)$), results in a hyperbolic solution. The dispersive factor arises from taking into account the dispersive media where mobile charges have a high probability of encountering deep traps with a slow release time which compete with other characteristic transport times (diffusion, injection/transfer). For a non-dispersive random walk, the number of sites visited by the mobile charges is linear ($S(t) \propto t$). On the other, considering a dispersion in the spatial separation between the localized states and a dispersion in the potential barriers between these sites, Scher and Montroll demonstrated that the waiting time distribution function could be well-represented by a power law $S(t) \propto t^{\alpha}$ where $\alpha = T/T_0$ and T_0 is the characteristic temperature of the distribution.[1,2] In the limit where $T_0 = T$ (or $\alpha = 1$), the distribution is inexistent and the solution is non-dispersive.

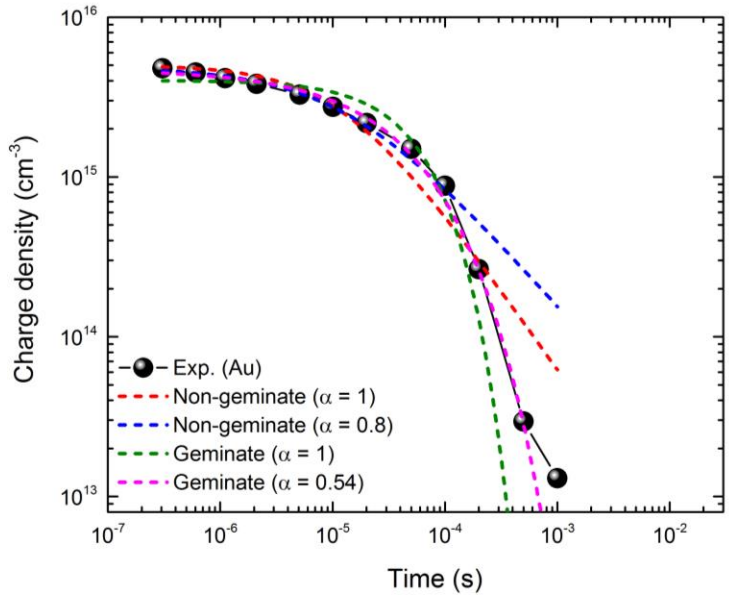

Figure S2. Experimental time-resolved charge density decay of the pixel without selective contact along with various fitting functions.

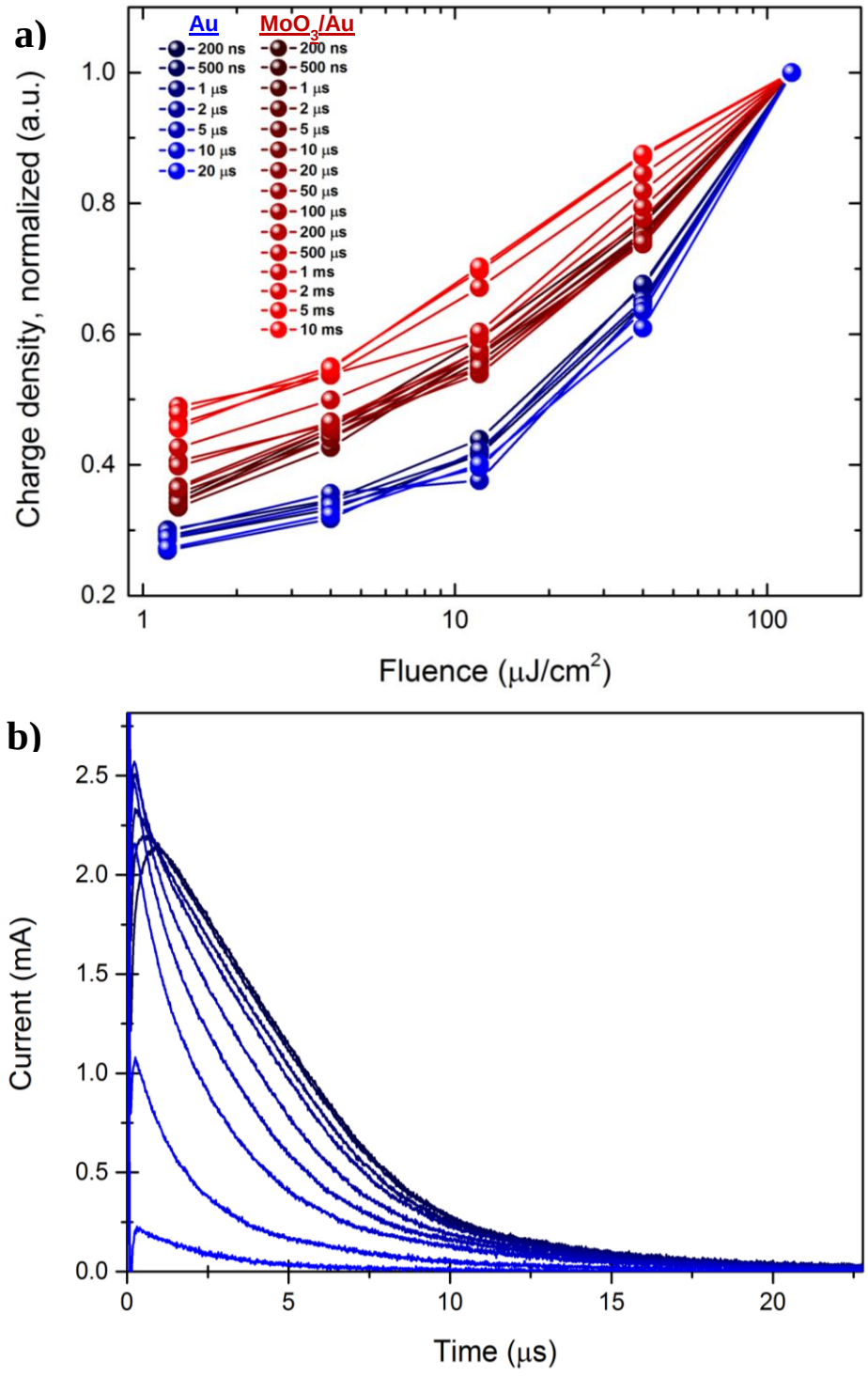

Figure S3. a) Comparison between normalized charge density in function of the light intensity, highlighting the fact that the decays are similar up to 20 µs. b) Current extraction curves for the device without selective contact under 40 µJ/cm^2 at various delay time (from 200 ns to 100 µs) highlighting the linear nature of the decay at fast delays.

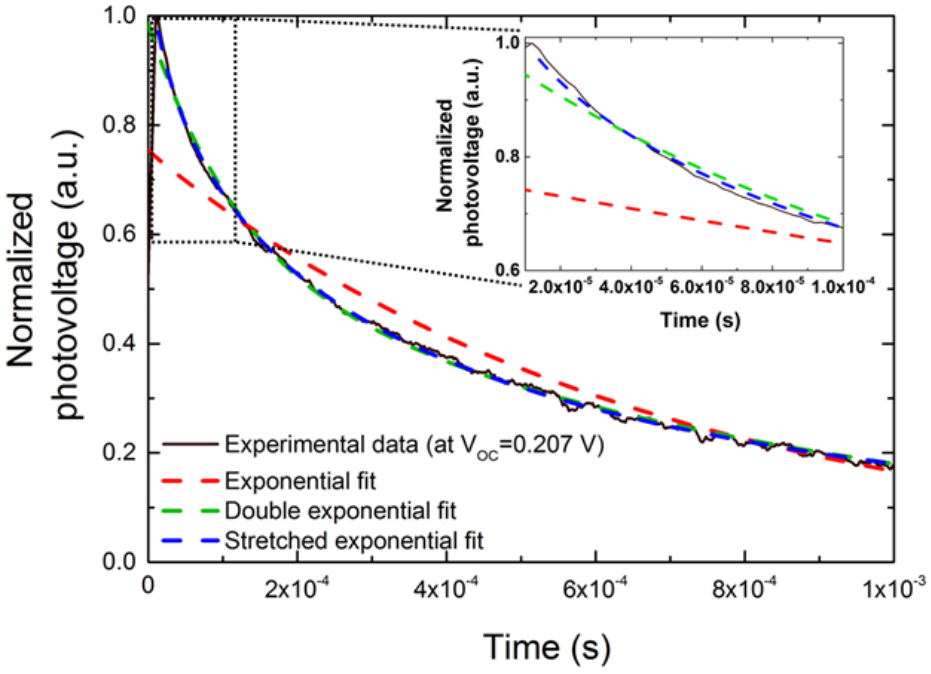

Figure S4. Typical TPV signal fitted with various exponential functions. Inset: high resolution of the early stage of the decay, highlighting the difference between the double exponential and the stretched exponential.

References

S1. Scher H.; Montroll E. W. Anomalous transit-time dispersion in amorphous solids. Phys. Rev. B 1975, 12, 2455-2477.

S2. Nelson J.; Chandler R. E. Random walk models of charge transfer and transport in dye sensitized systems. Coord. Chem. Rev. 2004, 248, 1181-1194.