Schottky Defects Suppress Nonradiative Recombination in $CH_3NH_3PbI_3$ through Charge Localization

Lu Qiao[1], Andrey S. Vasenko[2], Evgueni V. Chulkov[3], Run Long[1]

[1]College of Chemistry, Key Laboratory of Theoretical & Computational Photochemistry of Ministry of Education, Beijing Normal University, Beijing 100875, China
[2]HSE University, 101000 Moscow, Russia
[3]Donostia International Physics Center (DIPC), 20018 San Sebastián-Donostia, Euskadi, Spain

Hybrid organic-inorganic perovskites (HOIPs) are of great interest in recent years because their excellent optoelectronic properties enable high power conversion efficiencies in solar cells with the latest record of 26.7% [1]. Further improving the performance of HOIP solar cells requires a better understanding of the energy loss mechanisms. Structural defects usually serve as carrier recombination centers to accelerate the energy dissipation in HOIPs. In this report, we demonstrate some stoichiometric Schottky defects [2], specifically, PbI_2 and CH_3NH_3I vacancies, can actually suppress the nonradiative electron-hole recombination in $CH_3NH_3PbI_3$ [3]. We employed density functional theory and nonadiabatic molecular dynamics to simulate the material properties and carrier recombination dynamics. These Schottky defects rarely affect the bandgap of $CH_3NH_3PbI_3$, and no mid-gap states are introduced. However, the spatial distribution of holes is dramatically modified, reducing their overlap with electrons and reducing the corresponding nonadiabatic couplings. Especially, the holes are localized around the vacancies with enhanced structural distortions. Our simulations indicate the carrier lifetime is efficiently extended from 1 ns to 2.1 and 2.6 ns by PbI_2 and CH_3NH_3I vacancies, respectively, see **Fig. 1** [3]. This work suggests the counterintuitive potential of Schottky defects in enhancing the performance of HOIP solar cells.

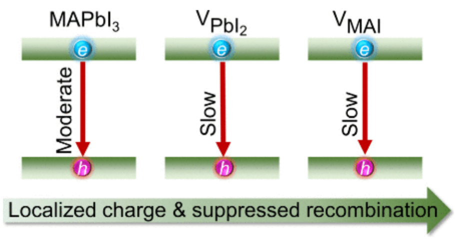

Fig. 1: *Scheme of Schottky defects extending the carrier lifetime in $CH_3NH_3PbI_3$. Here MA is $CH_3NH_3^+$.*

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References

1. M.A. Green, E.D. Dunlop, M. Yoshita, N. Kopidakis, K. Bothe, G. Siefer, D. Hinken, M. Rauer, J. Hohl-Ebinger, X. Hao, Solar Cell Efficiency Tables **32**, 425−441 (2024).
2. C. Ran, J. Xu, W. Gao, C. Huang, S. Dou, Chem. Soc. Rev. **47**, 4581−4610 (2018).
3. L. Qiao, A.S. Vasenko, E.V. Chulkov, R. Long, J. Phys. Chem. Lett. **16**, 1, 215-221 (2025).