Intraband Transitions Induced by Below-Bandgap Photoexcitation at $CsPbBr_3$/GaAs Heterointerface

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Two-step photon upconversion (TPU) is a physical phenomenon associated with the sequential excitation of an electron. Firstly, the electron is excited by a process called interband excitation (band-to-band excitation). The excited electron, then, absorbs a sub-bandgap photon and is excited to a higher energy state. The latter process is called intraband excitation. Two-step photon upconversion solar cells (TPU-SCs) were realized using III-V semiconductors, namely $Al_{0.3}Ga_{0.7}As$ and GaAs [1]. Since the semiconductors exhibit different energy bandgaps, the band discontinuity at the heterointerface (HI) allows carrier accumulation. In this case, electrons can be generated by the interband excitation in GaAs. The accumulated electrons at the HI can absorb sub-bandgap photons which allows the intraband excitation. The accumulating electron density can be enhanced by the incorporation of quantum structures which relax the optical selection rule. In this work, we investigate perovskite/III-V semiconductor HI and its influence on intraband excitation which can be improved further for TPU-SC applications.

We fabricate a simple inverted solar cell structure of Ag/ZnO/$CsPbBr_3$/p-GaAs/Au-Zn/Au. The $CsPbBr_3$ perovskite layer was grown by multi-step spin coating. Considering the achieved structure, $CsPbBr_3$/GaAs HI exhibits band discontinuity with energy difference at the conduction band and the valence band (denoted by ΔE_{CB} and ΔE_{VB} respectively) of $\Delta E_{CB} = 0.77$ eV and $\Delta E_{VB} = 0.11$ eV. The ΔE_{CB}:ΔE_{VB} ratio of 7:1 shows a higher theoretical energy conversion efficiency than $Al_{0.3}Ga_{0.7}As$/GaAs HI for which the ΔE_{CB}:ΔE_{VB} is approximately equal to 3:2 [2]. Therefore, $CsPbBr_3$/GaAs-based TPU-SCs can achieve higher efficiency than III-V semiconductor-based TPU-SCs.

We characterized the $CsPbBr_3$/GaAs-based TPU-SCs by various measurements. The External quantum efficiency spectrum measured under single-color conditions, i.e., only the interband excitation occurs, shows clear absorption band edges of $CsPbBr_3$ and GaAs. The measurement of gains in photocurrent and photovoltage (denoted by ΔJ_{SC} and ΔV_{OC} respectively) exhibit notable features. We measured the photocurrent and photovoltage under the 784-nm photoexcitation with and without additional 1319-nm sub-bandgap photoexcitation (two-color and single-color conditions respectively). The ΔJ_{SC} and ΔV_{OC} can be defined by the following:

$$\Delta J_{SC} = J_{SC,\ \text{two-color}} - J_{SC,\ \text{single-color}}$$

$$\Delta V_{OC} = V_{OC,\ \text{two-color}} - V_{OC,\ \text{single-color}}$$

The results show that ΔJ_{SC} increases with increasing 1319-nm excitation intensity. However, the increase in ΔJ_{SC} can be due to thermal activation at the HI. Here, ΔV_{OC} is an important indicator to distinguish the intraband excitation from the thermal activation. Since thermal carrier population induces a reduction in V_{OC}, ΔV_{OC} should be negative with increasing temperature. In contrast, the intraband excitation relates to the increase in electron density at the CB of the WGS (in this case, $CsPbBr_3$). Therefore, the observed positive values and increasing features of ΔV_{OC} with 1319-nm sub-bandgap photon intensity are strong evidence for the intraband excitation at the HI.

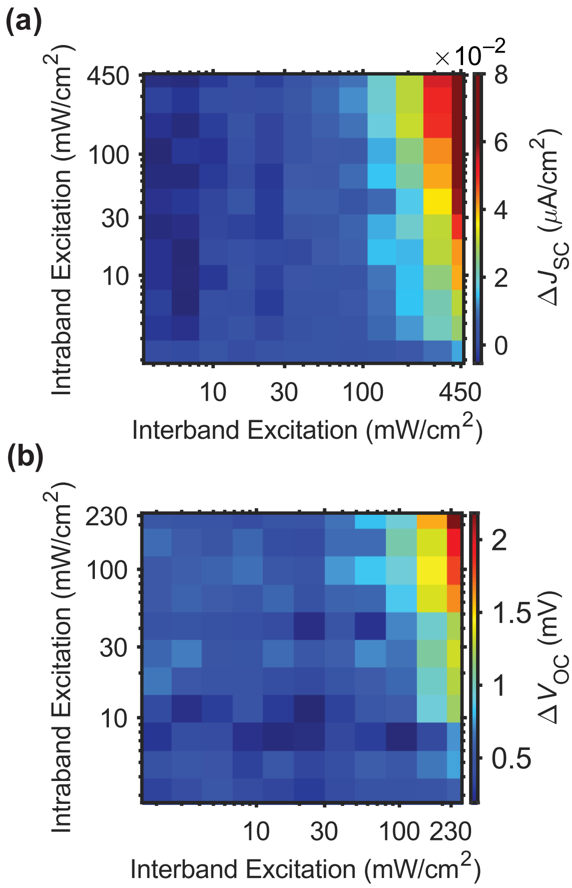

Fig. 1 (a) ΔJ_{SC} and (b) ΔV_{OC} maps visualized as functions of interband and intraband excitations

References

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[2] S. Asahi, K. Kusaki, Y. Harada, T. Kita, Sci. Rep. **8**, 872 (2018).