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## On the role of molecular charge transfer states in ultrafast photo-charge generation in solid-state dye sensitized solar cells

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The role of intermolecular charge transfer (CT) states in solid-state dye sensitized solar cells employing a perylene dye and metal-oxide electrodes are investigated comprehensively employing (a) ultrafast transient absorption, (b) transient photoluminescence and (c) optical pump-THz probe spectroscopy techniques. Here we show that the presence of a molecular excited state with a strong charge-transfer character may be critical for charge generation when the total energy of the photoexcitation is too low to intercept accepting states in the TiO<sub>2</sub> photoanode. Though hole transfer to the 2,2',7,7'-tetrakis(N,N-di-p-methoxyphenylamine)-9,9'-spirobifluorene (Spiro-OMeTAD) can be very fast, an electron - hole pair is likely to form at the organic interface, resulting in a possible trap like excitation. This leads to poor photocurrent generation in the solid state DSSC (ss-DSSC) device. Combining the information obtained from the three complementary ultrafast spectroscopy techniques mentioned above, we find that the molecular exciton is stabilized in a low-energy CT state through efficient intramolecular electron transfer and if the metal oxide presents a significant energy barrier, the photoexcited state may remain trapped at the organic interface, though hole transfer from the dye to the Spiro-OMeTAD happens efficiently. This will lead to a loss in charge generation and hence low photo-current. We demonstrate that it is possible to overcome this issue by fabricating SnO<sub>2</sub>-based ss-DSSC and performing similar spectroscopic analysis. We corroborate the above observation by fabricating and testing devices with both TiO<sub>2</sub> and SnO<sub>2</sub> photoanodes.

**主催:九州大学 最先端有機光エレクトロニクス研究センター**

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