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STUDY OF CHARGE CARRIER DYNAMICS IN ANATASE AND RUTILE TIO2: IMPLICATIONS ON PHOTOCATALYTIC WATER SPLITTING

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Understanding of the fundamentals behind charge carrier dynamics of photocatalytic materials is still illusive, hindering progress in our quest for renewable energy. TiO2 anatase and rutile are the most understood phases in photocatalysis and serve as the best model systems for fundamental studies. The ultrafast charge carrier dynamics, especially on TiO2 anatase single crystals (the most active phase), are unresolved. The femtosecond time-resolved spectroscopy was carried out to explore the dynamics of photoexcited charge carriers' recombination in the anatase single crystal, for the first time using pump fluence effects, and we compared it to that in the rutile single crystal. A significant difference in charge carrier recombination rates between both crystals is recorded. More specifically, we found that the time constants for carrier recombination are two orders of magnitude slower for anatase (101) when compared to those of rutile (110). Moreover, bulk defects introduced by reduction of the samples via annealing in ultrahigh vacuum resulted in faster recombination rates for both polymorphs. Both states (fresh and reduced) probed by pump fluence dependence measurements revealed that the major recombination channel in fresh and reduced anatase and reduced rutile is first-order Shockley–Read–Hall-mediated. However, for fresh rutile, third-body Auger recombination was observed and attributed to the presence of higher density of intrinsic charge carriers. At all excitation wavelengths and fluence investigated, the anatase (101) single crystal shows longer charge carrier lifetimes when compared to the rutile (110) single crystal. This may explain the superiority of the anatase phase than the rutile phase in M/TiO2 catalysts for molecular hydrogen production.

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