

## Probing the role of crystallinity in charge carrier dynamics of TiO<sub>2</sub> films using ultrafast transient absorption spectroscopy.

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The structural order of titanium dioxide (TiO<sub>2</sub>) films critically influences its photophysical properties, particularly charge carrier generation, trapping, and recombination processes. In this work, we examine how varying degrees of crystallinity affect the ultrafast charge carrier dynamics in TiO<sub>2</sub> thin films, employing transient absorption spectroscopy in the femtosecond to picosecond timescale. Films were subjected to different thermal treatments (from 300 °C to 550 °C) to systematically tune their crystallinity, ranging from predominantly amorphous to anatase phase. Structural characterization indicates an increase in crystallite size with calcination temperature, reflecting enhanced structural order within the TiO<sub>2</sub> framework. Transient absorption measurements at 325 nm excitation, performed under multiple pump fluences (100–3000 μJ·cm<sup>-2</sup>), reveal distinct differences in carrier dynamics as a function of crystallinity. Amorphous samples display rapid signal decay, consistent with a high density of trap-assisted recombination pathways. In contrast, films treated at higher temperatures exhibit slower decay components, indicative of extended carrier lifetimes and reduced recombination rates. These results suggest that improved crystallinity reduces defect-related trapping and facilitates more efficient charge transport within the TiO<sub>2</sub> network. Furthermore, excitation density-dependent measurements reveal that crystalline samples are more dependent on excitation power, exhibiting the fastest decay times with increasing pump power. Overall, this work demonstrates that crystallinity is a key parameter governing charge carrier dynamic in TiO<sub>2</sub> films, providing valuable insights for tailoring materials for photoactive applications.