Long-lived Photo-Response of Multi-Layer N-Doped Graphene-based Films

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The future of solar energy storage depends on affordable and efficient solutions in the form of solar chemicals.[1,2] Therefore, it has become necessary to study solar chemical production from earth abundant raw materials such graphene-based films produced from the pyrolysis of biomass like chitosan.[3,4] These doped or defective graphenes have been shown by several researchers to exhibit improved photo (electro) catalysis compared to pristine graphene.[5,6] Moreover, they are strong candidates to replace options such ITO.[7]

However, the applications of doped graphene are limited by a lack of knowledge on the underlying mechanisms responsible for their photo (electro) catalysis.[6] Without this, doping strategies can be inefficient or even counterproductive at increasing solar fuel production.[8] For efficient solar fuel production, the photo-generated electrons must be available for a sufficiently long time to migrate to the photoelectrode surface for subsequent interfacial reactions.[6] However, to the best of our knowledge, all reported ultrafast laser spectroscopic measurements had only revealed a sub-picosecond lifetime of photocarriers in doped graphene,[9,10] which is too short to initiate photo (electro) catalytic reactions efficiently.

We undertook a more careful transient absorption study of doped graphene in longer time scales and observed a weak but long-lasting response extended to ns time domain. The transient transmittance and reflectance responses of N-doped graphene samples on quartz substrate prepared by chitosan pyrolysis at 900–1200°C compared to an undoped graphene control were explored. These responses were recalculated as (complex) dielectric function changes and decomposed into Drude-Lorentz parameters, to determine their origin. All samples had an expected photo-response; fast relaxation (within 1 ps) due to decreased plasmon damping and increased conductivity. But the N-doped graphenes had an additional transient absorption signal of 10x lower intensity with lifetime extending into the ns domain. This long-lived response was attributed to carrier trapping at N-doping centres, which presumably contributes to the improved catalytic activity of doped graphenes.[11]

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