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Iron pyrite nanocubes as photon absorbers: Unraveling the issues

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Iron pyrite (FeS2) has a high absorption coefficient and a suitable bandgap for photovoltaic applications but its poor performance in experimental devices has precluded its use. This is loosely attributed to impurity phases and defects but definite proofs have not been forthcoming. For successful use in a photovoltaic device, the extraction of photoexcited carriers without loss from the absorber material is a key requirement. With the advent of chemical synthesis techniques for nanoparticles, it is now possible to produce pure pyrite without impurities. In this paper, we deal with fundamental photophysics and possible carrier loss mechanisms in films made from pure pyrite nanocubes synthesized by a hot injection method. The nanocubes are (100) faceted. The optical, transport and magnetic properties are evaluated. Ultrafast transient absorption spectroscopy was done to elucidate the charge carrier lifetimes during relaxation. We found fast carrier localization of photoexcited charges to indirect band edge and shallow trap states with a short characteristic decay time, followed by relaxation to deep states and recombination of trapped carriers with long characteristic decay times. Temperature dependence of electrical resistivity exhibits a Mott-variable range hopping (VRH) conduction mechanism consistent with the presence of high density of defects. Temperature dependence of magnetization showed magnetic ordering at low temperatures which could be associated with sulfur vacancies clustering when the thermal vibrations are low to give rise to magnetically ordered, sulfur deficient phases such as $Fe_{\delta}S_{1-\delta}$ ($0 \leq \delta \leq 1$).

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