

The bandgap of pyrite: Bandgap oscillations on an ultrafast timescale

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Iron pyrite (fool's gold) has attracted attention as a potential solar cell material because of its good bandgap, high absorption, low cost, abundance, and environmentally benign components. Despite its high promise, photoelectrochemical cells made from pyrite typically exhibit poor performance. Although much work has been undertaken to understand the properties of pyrite, little attention has been paid to its dynamical properties. The conduction band minimum of pyrite is comprised of sulphur p σ^* anti-bonding orbitals so its energy depends critically on the sulphur-sulphur distance. As there are a number of phonon modes that dynamically change this distance, the bandgap of pyrite is a dynamical quantity. In particular, there is a mode at 347 cm^{-1} that consists of sulphur atoms oscillating with respect to one another. This sets up bandgap oscillations on a 100 fs timescale. The magnitude of these oscillations is temperature dependent but they can be extremely wide, taking the material from a near metal to a wide bandgap semiconductor with even a modest change in the sulphur-sulphur distance. In this work, we use fully self-consistent GW calculations to elucidate the band structure of pyrite and to track its changes upon excitation of phonons in the material.