

Abstract Submitted  
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**The effect of surface stoichiometry on the band gap of the pyrite  $\text{FeS}_2(100)$  surface**<sup>1</sup> YANNING ZHANG, JUN HU, MATT LAW, RUQIAN WU, University of California, Irvine — Iron pyrite ( $\text{FeS}_2$ ) is experiencing a resurgence of interest for use in solar photovoltaic and photoelectrochemical cells. The main hurdle to the use of pyrite is the low open-circuit voltage of pyrite devices, which may result from gap states created by surface and bulk defects. Recently, systematic spin-polarized DFT calculations were performed for a series of pyrite  $\text{FeS}_2(100)$  surfaces to clarify the effect of surface stoichiometry on stability, electronic structure, and band gap. It was found that while stoichiometric and S-poor  $\text{FeS}_2(100)$  surfaces are semiconductors with band gaps of 0.56-0.72 eV, S-rich surfaces are small-gap semiconductors ( $E_g < 0.3$  eV) or metals. The stoichiometric  $\text{FeS}_2(100)$  surface is spin polarized in the topmost layer ( $2 \mu_B$  per Fe) and displays a band of Fe  $d_{z^2}$  gap states centered 0.2 eV above the valence band edge. Our calculations suggest that the low open-circuit voltage of pyrite solar cells may result from a narrowed surface band gap. S-poor surfaces may provide larger photovoltages than S-rich surfaces. The segregation process of sulfur vacancy under different surface conditions are also being studied, so as to provide useful guidelines for the design and fabrication of better pyrite photovoltaic materials and devices.

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