

Scanlon and Watson Reply: In our recent Letter [1], we demonstrated using screened hybrid density functional theory that H *does not* act as a shallow donor in *p*-type Cu_2O , which is opposite to the behavior of H in many *n*-type oxides. Furthermore, we demonstrated that H will impede *p*-type conductivity in Cu_2O , and proposed strategies to remove H from Cu_2O samples, and hopefully increase conductivity.

In their Comment [2], Biswas *et al.* have questioned our description of the H_i^{tet} as being “quasiatomic,” stating that H is anionic in the H_i^{tet} position, in all three charge states. We have never argued that the H atom is not anionic, or that the charge state of H changes in the three different charge states, as Biswas *et al.* seem to indicate. We stated the H_i^{tet} was quasiatomic, with quasiatomic in this instance being defined as per Refs. [3,4] as suggesting: “considerable overlap of the muonium orbital onto adjacent Cu^+ ions.” This is clearly evidenced by Fig. 2 in the original Letter, [1]. We did not state that H_i^{tet} was “quasiatomic with multiple charge states” as Biswas *et al.* have claimed, we simply reported that the H_i^{tet} was quasiatomic and showed that the defect was amphoteric, which Biswas *et al.* have also found [2]. We agree with Biswas *et al.* that H is anionic in all three charge states, but this does not make our description of the H_i^{tet} incorrect in any way. In fact, the results of Biswas *et al.* simply serve to confirm our analysis of H_i^{tet} being an amphoteric defect, and showing that interstitial H cannot act as a donor in Cu_2O .

Biswas *et al.* also questioned the charge states we observe for the H- V_{Cu} complex, stating that the neutral H- V_{Cu} should possess no defect states in the band gap, and therefore should not be amphoteric. In their calculations, they do not observe the deep state that we observe for the H- V_{Cu}^{-1} , although in the Comment they incorrectly state that we see the deep level for the H- V_{Cu} neutral state. We agree, the neutral H- V_{Cu} does not possess any deep levels in the band gap, and we have never claimed that it did. We reported that the 0/−1 ionization level was mediated by a singly occupied single particle level, present from the −1 charge state. We suggested this ionization level was the source of the ionization level reported in the muon spectroscopy experiments[3]. Nevertheless, whether the H- V_{Cu} defect possesses ionizations levels or not, the formation energy of the complex is extremely low, and indicates that H will bind into any V_{Cu} in the system, retarding *p*-type conductivity.

Lastly Biswas *et al.* have questioned our findings that H_O is an amphoteric defect. They find that H acts as a shallow donor, which they say is in agreement with the behavior of H in other oxides, and at variance with our results. Shallow donor behavior of H substituting for an O

is found in *n*-type oxides such as SnO_2 [5], In_2O_3 [5], CdO [6], and ZnO [7]. All these materials, however, are characterized by low conduction band minima relative to the vacuum level. H_O has never been shown experimentally to act as a shallow donor for materials with a conduction band minimum (CBM) significantly higher than ~ -4.5 eV relative to the vacuum. The CBM of Cu_2O lies at ~ -3.5 eV relative to the vacuum level (from the most recent experimental measurements [8]), and thus H_O would not be expected to act as a shallow donor in this system. Even if H could act as a shallow donor in Cu_2O , the formation energy at the CBM is prohibitively high, and at higher Fermi levels, the H_i^{tet} in the −1 charge state will compensate it, killing any *n*-type conductivity.

In conclusion, the results of Biswas *et al.* [2], and our own results [1], serve to show that H cannot act as an effective shallow donor in Cu_2O , and that H will bind strongly into any V_{Cu} in the system, killing the native *p*-type conductivity.

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Received 11 April 2012; published 23 May 2012

DOI: 10.1103/PhysRevLett.108.219704

PACS numbers: 71.20.Nr, 71.55.−i, 84.60.Jt

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