Agoston, Peter; Albe, Karsten; Nieminen, Risto M.; Puska, Martti

Comment on "Intrinsic n-type Behavior in Transparent Conducting Oxides"

Published in: Physical Review Letters

DOI: 10.1103/PhysRevLett.106.069602

Published: 10/02/2011

Please cite the original version:

This material is protected by copyright and other intellectual property rights, and duplication or sale of all or part of any of the repository collections is not permitted, except that material may be duplicated by you for your research use or educational purposes in electronic or print form. You must obtain permission for any other use. Electronic or print copies may not be offered, whether for sale or otherwise to anyone who is not an authorised user.
Ágoston et al. Reply: In a recent work we studied the oxygen vacancy in \(\text{In}_2\text{O}_3\), \(\text{SnO}_2\), and \(\text{ZnO}\) by means of hybrid-functional calculations within density functional theory [1]. In particular, our interpretation of the intrinsic electron donation mechanism in the transparent conducting oxide \(\text{In}_2\text{O}_3\) has been questioned by Lany and Zunger [2]. In the following we reply to their comments.

Lany and Zunger argue that our results indicate a deep color center behavior of the oxygen vacancy in \(\text{In}_2\text{O}_3\), because the defect state of a shallow defect should be resonant well above the conduction band minimum as in extrinsically doped transparent conducting oxides (TCO). In contrast to extrinsically doped TCOs like \(\text{SnO}_2:\text{F}\) or \(\text{In}_2\text{O}_3:\text{Sn}\), the oxygen vacancy state does in fact produce a much deeper state which, however, may still allow a thermal ionization of the defect if the subsequent structural relaxation of the defect provides a sufficient energy gain in the system.

While we find that the oxygen vacancy is a shallow donor in \(\text{In}_2\text{O}_3\) we did not claim that the \(V_O\) is able to produce a pronounced Burnstein-Moss shift. This effect has mostly been observed for extrinsically doped TCOs. Moreover, the occurrence of a shift in nominally undoped \(\text{In}_2\text{O}_3\) together with measured electron concentrations \(>10^{20}\ \text{cm}^{-3}\) [3] cannot be explained by the oxygen vacancies alone. We believe that such effects are likely to be produced by hydrogen defects which has recently been suggested also for \(\text{In}_2\text{O}_3\) [4].

According to Lany and Zunger, it is the “biased” choice of the range separation parameters of our hybrid density functional calculations which results in equilibrium transitions close to the conduction band minimum. We have adopted the range separation parameters for the different materials which reproduce the experimentally observed trends for the size of the band gaps going from \(\text{SnO}_2\) via \(\text{In}_2\text{O}_3\) to \(\text{ZnO}\), which cannot be reproduced by a single hybrid functional. Moreover, in the case of \(\text{In}_2\text{O}_3\) the actual value of the band gap is still a matter of debate and the published data span a range from 2.67 eV to maximally 3.1 eV [5–7]. The additional results which Lany and Zunger have provided in their comment and using quasiparticle corrections leads undoubtedly to the conclusion that the ionization energy of the vacancy should be increased in that case. We note, however, that the final conclusion still depends on the actual choice of the band gap.

Péter Ágoston* and Karsten Albe
Institut für Materialwissenschaft
TU Darmstadt, Petersenstr. 23
D-64287 Darmstadt, Germany

Risto M. Nieminen and Martti J. Puska
Department of Applied Physics
Aalto University, P.O. Box 11100
FIN-00076 AALTO, Finland

Received 9 April 2010; published 10 February 2011
DOI: 10.1103/PhysRevLett.106.069602
PACS numbers: 61.72.Bb, 61.72.jd, 71.15.Nc, 76.30.Mi

*agoston@mm.tu-darmstadt.de