Low Hole Effective Mass $p$-type Transparent Conducting Oxides:
Identification and Design Principles

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Abstract

The development of high performance transparent conducting oxides (TCOs) is critical to many technologies from transparent electronics to solar cells. While $n$-type TCOs are present in many devices, current $p$-type TCOs are not largely commercialized as they exhibit much lower carrier mobilities, due to the large hole effective masses of most oxides. Here, we conduct a high-throughput computational search on thousands of binary and ternary oxides and identify several highly promising compounds displaying exceptionally low hole effective masses (up to an order of magnitude lower than state of the art $p$-type TCOs) as well as wide band gaps. In addition to the discovery of specific compounds, the chemical rationalization of our findings opens new directions, beyond current Cu-based chemistries, for the design and development of future $p$-type TCOs.
Transparent conducting oxides (TCOs) are compounds exhibiting high electrical conductivity and transparency to visible light. Those materials are needed in many applications from solar cells, where a TCO thin film provides electrical contact without impeding the flux of visible light reaching the device, to transparent transistors that could, for instance, be integrated in windows.[1–5] The main strategy to achieve the two antagonistic properties of high conductivity and transparency is to use wide band gap oxides (favoring transparency) doped with a significant amount of mobile charge carriers, either holes (\( p \)-type) or electrons (\( n \)-type).[6] \( n \)-TCOs (e.g., indium tin oxide, ITO) are already present in many modern devices but \( p \)-TCOs have not been largely commercialized as their carrier mobilities stand an order of magnitude behind their \( n \)-counterparts. This situation impedes many critical technological developments from more efficient organic and thin film solar cell designs, benefiting from a better band matching by using \( p \)- instead of \( n \)-TCOs,[7, 8] to the entire new field of transparent electronics which requires both \( p \)- and \( n \)-type TCO materials.[2, 9–13]

There is a fundamental reason to the difficulty of developing high mobility \( p \)-type TCOs: the localized oxygen \( p \) nature of the valence band in most oxides makes those bands very flat and leads to large hole effective masses.[12, 13] The field of \( p \)-type TCOs received most of its impulse a decade ago when Kawazoe et al.
demonstrated that CuAlO\(_2\) delafossite could show encouraging \( p \)-type conductivity and optical transparency in the visible.[14] The unusual hole mobility of CuAlO\(_2\) was explained by a large hybridization of the oxygen orbitals with \( 3d^{10} \) electrons in the Cu\(^{1+}\) closed shell, lowering the oxygen character and leading to dispersive (low effective mass) valence band. This finding led to the outline of a design rule for \( p \)-type TCOs requiring the presence of Cu\(^{1+}\) and motivated the study of a very large range of Cu-based materials [12, 13] such as other delafossites (e.g., CuCrO\(_2\),[15]), SrCu\(_2\)O\(_2\),[16] or Cu-based oxychalcogenides.[18]

To this day, the question remains open wether alternative chemistries and design rules could lead to materials with lower hole effective mass. Answering this question is critical for the \( p \)-type TCO field as it would enable the identification of the high hole mobility oxides that the TCO community has been looking for. Traditionally, design principles are developed by the rationalization of experimentally observed data. In this work, we take an alternative path using a database of high-throughput \textit{ab initio} computed data containing electronic structure for thousands of binary and ternary oxides.[19–22] By browsing this database, we identify the compounds and chemistries leading to low hole
effective masses. This enables us to uncover the underlying chemical reasons for low hole effective masses and to propose novel and unsuspected design rules for the development of $p$-type TCOs.

**Results**

**Hole vs electron effective mass distribution in oxides**

Our database contains density functional theory (DFT) band structures for 3052 oxides. All the oxides that we have studied are *existing* minerals, or *already synthetized* materials, whose experimentally measured crystalline structure has been taken from the ICSD database. We have taken their first principles relaxed crystalline structure as available in the Materials Project Database, and computed their electronic structure (band gaps and effective masses) using state of the art methodologies, as described in the methods section and in supplementary information. Figure 1 shows the histogram of holes (in red) and electrons (in blue) effective mass. The difference in distribution between hole and electron effective mass compounds is striking, emphasizing that finding high mobility $p$-type oxides is indeed significantly more challenging than for $n$-types. The chemical reasons for such a difference comes from the very different character of the valence and conduction bands in oxides. The valence bands tend to be of oxygen $p$ localized character (leading to large effective masses), while the conduction bands are cationic and present more often dispersive bands (i.e., low effective mass).
Figure 1: Histogram of the maximum line effective mass for holes (valence band) in red and electrons (conduction band) in blue in our set of binary and ternary oxides. The effective mass bin size is 0.2 and the figure focuses on the region of low effective mass (lower than 5).

**High-throughput identification of low hole effective mass, wide band gap oxides**

While low hole effective mass oxides are rare, our large database gives us the opportunity to identify the outliers. By filtering for oxides with an effective mass lower than 1.5, we end up with only 20 candidates that have never been considered as TCOs. Details on the screening procedure and results are available in methods and supplementary information. To further assess the practical interest of those candidates, we also evaluate their band gaps as this would influence their light absorption. As DFT is known to not as accurately model band gaps than band shape and width, we computed band gaps with a higher order method: many body perturbation theory in the $GW$ approach.\textsuperscript{[28, 29]} Figure 2 plots the effective mass vs the band gap for the 20 identified compounds (red dots). For comparison, we also plotted a few known $p$-type TCOs (blue diamonds, Cu-based TCOs and ZnRh$_2$O$_4$) and some current $n$-type materials (green squares, SnO$_2$, In$_2$O$_3$, ZnO). The compounds we identified beat the state of the art $p$-type TCOs by up to an order of magnitude in effective mass. Some compounds are even reaching hole effective mass values close to the best electron effective mass in oxides exhibited by current $n$-type materials. This shows that the large difference in mobility between current $n$-type and $p$-type materials is not inevitable and could be overcome by the investigation of alternative
The best TCOs should lie in the lower right corner as they would show small effective masses and large band gaps. However, the materials containing toxic elements such as Pb, Tl and Hg are less likely to be of interest technologically. The series of $A_4B_2O$ oxypnicnitudes compounds ($A = \text{Ca, Sr and } B = \text{P, As}$) and NaNbO$_2$ are not of high priority as compounds with both higher band gaps and lower effective masses are present in our data set. K$_2$Sn$_2$O$_3$ shows the lowest effective mass around 0.27-0.28 but a band gap on the small side (2.4 eV). Interestingly, the band gap of K$_2$Sn$_2$O$_3$ can be increased by substituting Na which would lead to lower absorption in the visible (see supplementary information). On the other hand, the ZrOS and HfOS compounds have larger effective masses but with a significantly larger band gap and opportunities for full visible range transmission. Finally, both Sb$_4$Cl$_2$O$_5$ and B$_6$O stand as the compounds of major interest with both large band gaps (respectively 3.6 and 3 eV) and low effective masses (respectively 0.37 and 0.59), while made of non-toxic, abundant elements.

**p-type dopability in the low hole effective mass, wide band gap candidates**

Effective masses and band gaps are necessary conditions for good p-type TCOs but they are not sufficient. One key requirement for a p-type TCO is to be able to be doped (intrinsically or extrinsically) with acceptor defects that will generate holes in the valence band. It is well reported that certain oxides are intrinsically difficult to dope p. The easy formation of compensating intrinsic defects (hole-killers) as the oxygen vacancy when lowering the Fermi energy towards the valence has been a main issue for the p-doping of certain oxides. Doping studies for other applications than TCOs have already been reported for several of the chemistries we identified. B$_6$O has been experimentally measured to be p-type, PbTiO$_3$ has been identified as p-type dopable by computations, in agreement with experimental results on PbZr$_{0.5}$Ti$_{0.5}$O$_3$ and recently computations showed that to oxygen vacancy is not a hole-killer in ZrOS. For the remaining compounds of greatest interest, we performed defect computations for all the vacancy intrinsic defects (see supplementary information). From these results we can conclude that Sb$_4$Cl$_2$O$_5$ is not likely to be p-type doped due to strong hole compensation by oxygen and chlorine vacancies. On the other hand, K$_2$Pb$_2$O$_3$ as well as K$_2$Sn$_2$O$_3$ (in both polymorphs) shows an oxygen vacancy very high in energy and that will not compensate hole formation. Moreover, the presence of a low energy potassium vacancy could lead to intrinsic p-type
behavior for those materials.

![Figure 2: Effective mass vs band gap for the p-type TCO candidates (red dots). We superposed on the band gap axis a color spectrum corresponding to the wavelength associated with a photon energy. A few known p-type (blue diamonds) and n-type (green square) TCOs can be compared to the new candidates. The best TCOs should lie in the lower right corner.](image)

**Discussion**

We now turn to analyzing the chemical reasons for exceptionally low effective masses and propose guidelines for future TCO design. Figure 3 and ?? show the band structure of one representative of each of the chemistries identified (K$_2$Sn$_2$O$_3$, Ca$_4$P$_2$O$_7$, Tl$_4$V$_2$O$_7$, PbTiO$_3$, ZrSO, B$_6$O and Sb$_4$Cl$_2$O$_5$). The color scheme indicates the nature of the band obtained by projecting the wave functions on the different elements. For the ternary compounds, one of the red, green or blue color is associated with the different elements and the resulting color is obtained by mixing those primary colors proportionally to the projections. The red color is always associated with oxygen. For the only binary B$_6$O, red is used for oxygen and blue for boron. All compounds have dispersive valence bands, with a high curvature near the valence band maximum indicative of a low hole effective mass. For all of them, the oxygen character of the valence band is very mild in agreement with the
localized nature of oxygen 2\textit{p} orbitals. While our candidates cover different chemistries, the low effective masses valence bands can be explained by two main mechanisms which are both related to a chemical way of producing valence bands with low oxygen character.

The first mechanism leading to compounds with low effective masses and wide band gaps concerns K$_2$Sn$_2$O$_3$, Rb$_2$Sn$_2$O$_3$, PbTiO$_3$, PbZrO$_3$, PbHfO$_3$, K$_2$Pb$_2$O$_3$, Tl$_4$O$_3$ and Tl$_4$V$_2$O$_7$ and is illustrated in Figure 3. For these compounds, the low hole effective mass originates from the hybridization of the \textit{s} states of a (n-1)\textit{d}$_{10}$\textit{n}\textit{s}$_2$ cation (e.g. Sn$^{2+}$, Pb$^{2+}$, or Tl$^{1+}$) with oxygen 2\textit{p} orbitals. This is similar to what happens for Cu-based compounds where the closed 3\textit{d} shell is hybridized with oxygen. However, \textit{s} are significantly more delocalized than \textit{d} orbitals leading to higher dispersion of the valence band and lower effective masses.(refs Walsh) So far, the most studied (n-1)\textit{d}$_{10}$\textit{n}\textit{s}$_2$ ion \textit{p}-type oxide has been SnO.[30] However, SnO suffers from a very small indirect gap (0.7 eV)[31] and its valence band shows anisotropic effective mass due to its layered structure. Our work shows that going to ternary oxides of Sn$^{2+}$ can lead to wider band gaps (similarly than going from Cu$_2$O to CuAlO$_2$ widens the gap of Cu-based compounds) but also to more isotropic effective masses by modifying the crystal structure. The Hg$_2$SO$_4$ compound shows also an orbital overlap of \textit{s} electrons with oxygen \textit{p} but with a slightly different electronic configuration as Hg$^{1+}$ is 5\textit{d}$_{10}$6\textit{s}$_1$.

From our set of (n-1)\textit{d}$_{10}$\textit{n}\textit{s}$_2$-containing compounds, we found that the oxygen hybridization is the most pronounced for Sn$^{2+}$ followed by Pb$^{2+}$ and finally Tl$^{1+}$ which has the least dispersive valence bands and the strongest oxygen character. While Bi$^{3+}$ is also (n-1)\textit{d}$_{10}$\textit{n}\textit{s}$_2$ and some Bi-based compounds were close to meet our criteria (e.g., BiVO$_4$, Bi$_2$Ti$_4$O$_{11}$, and BiBrO, see supplementary information), none of them passed the 1.5 cut-off on average effective mass. It is possible that our data set did not contain the adequate crystal structure to lead to large bismuth \textit{s}-oxygen \textit{p} overlap in every directions but that such a structure might be achievable. A Sb$^{3+}$-based compound (Sb$^{3+}$ is 4\textit{d}$_{10}$ 5\textit{s}$_2$) is also present in our candidates (i.e., Sb$_4$Cl$_2$O$_5$). However, here not only does the \textit{s} orbital of Sb hybridize with oxygen but the valence band character is also influenced by the presence of anionic chlorine.
Figure 3: Band structures for a series of representative $p$-type TCOs identified in this work. The band structure is computed by GGA but a rigid shift of the conduction band (scissor operator) is applied to fit the band gap to the more accurate value obtained by $GW$. The color indicates the character of the bands by projections of the wave function on the different sites and orbitals. Each ternary compound has one of the red, green or blue color associated with it and the resulting color is obtained by mixing them in proportion equivalent to the projections. The red color is always associated with oxygen. For the only binary $B_6O$, we used red for oxygen and blue for boron.
The presence of an additional anion is actually the second mechanism at play in our low effective mass candidates (i.e., ZrOS, HfOS and the oxypicnitides) and presented in Figure 3. Here, the valence band keeps a $p$ character but more delocalized orbitals such as $3p$ (for $S^2-$, $P^{3-}$) or $4p$ ($As^{3+}$) replace the oxygen $2p$ valence band. Although, pure $3p$ or $4p$-orbital-based compounds have often too small band gaps, the mixing of oxygen and another $p$-orbital anion can lead to compounds with low effective masses and large band gaps. For instance, the band gaps of ZrS$_2$ is 1.7 eV$[32]$ while ZrO$_2$ shows a larger band gap (from 5.2 to 5.7 eV$[33]$) but flat oxygen valence bands. ZrOS achieves an interesting trade-off offering a larger band gap than for the sulfide with lower hole effective masses due to the sulfur character of the valence band. Our study did not lead to any oxypicnitides offering such a good trade-off but it is a still unexplored chemistry where future TCO work might be fruitful. Boron suboxide actually also fits in the anion mixing category as B$_6$O can be seen as a mixture of cationic boron, anionic boron and anionic oxygen.

From this analysis, two novel design principles can be outlined to achieve low hole effective masses oxygen containing compounds: the use of $(n-1)d^{10}ns^2$ ions especially Sn$^{2+}$ (but also Bi$^{3+}$ or Sb$^{3+}$) or the presence of another anion with $p$-orbitals more delocalized than oxygen (e.g., $S^2-$, $P^{3-}$, $Cl^-$ or $Br^-$). Both approaches can be combined as in Sb$_4$Cl$_2$O$_5$. Beyond the compounds we have already identified, there are still many opportunities for discovering new low hole effective mass TCOs (e.g., previously unknown ternaries or quaternaries) with the guidance of those design principles.

Motivated by the technological need for high mobility $p$-type TCOs and the difficulty to design such materials, we studied a large database of computed band structures for thousands of oxides, searching for the chemistries prone to low effective masses oxides. Our large scale study confirms that holes tend to have higher effective masses than electrons in oxides, but identifies several non-toxic, earth-abundant compounds with exceptionally low hole effective masses and wide band gaps: B$_6$O, Sb$_4$Cl$_2$O$_5$, $A_2Sn_2O_3$($A$ = K, Na) and ZrOS. By analyzing those results, two novel design principles, leading to compounds with lower effective masses than state of the art $p$-type TCOs, emerge: hybridization of a $(n-1)d^{10}ns^2$ with oxygen and/or mixed anionic systems. Those principles are extremely valuable to guide future searches for high mobility $p$-type TCO and, after a decade of exploration of Cu-based chemistries, they offer new chemical spaces full of promises.
Methods

All the high-throughput DFT computations have been performed using the Vienna ab initio software package (VASP),[34] with PAW pseudopotentials[35] and the generalized gradient approximation (GGA) as implemented by Perdew, Burke and Ernzerhof (PBE).[36] All ionic relaxations have been performed using AFLOW[37] and the high-throughput computations parameters are described in Jain et al.[25]

We considered all compounds containing less than 100 atoms in the unit cell present in the Materials Project Database originally from the Inorganic Crystal Structure Database (ICSD) and containing oxygen and no elements such as rare-earths (from Z=58, Ce to Z=71, Lu), inert gases, and any element with an atomic number larger than 84 (Po).

To perform the high-throughput band structure computations, we used the Materials Project structures (already relaxed with GGA) and performed a static run to obtain the charge density followed by a non-self-consistent band structure run along the band structure symmetry lines provided by Curtarolo et al.[20, 37] The full average effective mass tensor computation was performed on a regular gamma centered 8,000 \(k\)-points grid interpolated using the Boltztrap code.[38]

In the one-shot \(GW\) approach, corrections are obtained perturbatively from a starting DFT electronic structure. \(GW\) and preparatory DFT calculations on the 20 target compounds are performed with the ABINIT code[39] at optimized geometries, obtained from the Materials Project database. The exchange correlation energy for the preparatory DFT computation is described using the local density approximation (LDA) functional.[40] The Brillouin Zone in sampled with Monkhorst-Pack grids and the \(k\)-point sampling density is similar for all considered systems (> 450/\(n\) \(k\)-points where \(n\) is the number of atoms in the unit cell). For each oxide, the planewave cutoff is determined separately and set using a total energy difference convergence criterion, leading to electronic energies converged within 10\(^{-3}\) eV on average. We use norm-conserving pseudopotentials to model the electron-ion interaction (see supplementary information). If any, we include semi-core \(d\) states as valence in the pseudopotential. The \(GW\) calculations are carried out using the the well established Godby-Needs plasmon pole approximation.[41] We use a cutoff of 20 Rydberg for the expansion of the dielectric matrix and a total number of \(\sim\)1300 bands for all oxides.

All analysis of the data (e.g., line effective mass computations or band structure plotting) was performed using the pymatgen python package.[42]
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