Undercoordinated indium as an intrinsic electron-trap center in amorphous InGaZnO$_4$

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Undercoordinated indium (In*) is found to be an intrinsic defect that acts as a strong electron trap in amorphous InGaZnO$_4$. Conduction electrons couple with the under-coordinated In* via Coulomb attraction, which is the driving force for the formation of an In*-M (M = In, Ga, or Zn) bond. The new structure is stable in the electron-trapped (2−) charge state, and we designate it as an intrinsic (In*-M)$^{2−}$ center in amorphous InGaZnO$_4$. The (In*-M)$^{2−}$ centers are preferentially formed in heavily n-doped samples, resulting in a doping limit. They are also formed by electrical/optical stresses, which generate excited electrons, resulting in a metastable change in their electrical properties.

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INTRODUCTION

The identification of charge-trapping defects on the atomic scale has been achieved in crystalline semiconductors. A donor can capture carrier electrons with large lattice relaxations, forming a DX (donor) complex. Although [O−(M−O)−M] is the same, the defect properties appear to be different, depending on which term (V$_O$ or M$_i$) is used because, in crystalline oxides, V$_O$ and M$_i$ have quite different properties. Thus, the terminology used for defects in crystalline solids may not be appropriate for use in amorphous solids, leaving the true nature of defects in amorphous solids unclear. This argument is independent of the charge state of the defects. For [O−(M−M)−O], when one of the M’s is divalent (M$_{II}$), such as Zn, the equivalence is still valid for the (2+) charge state: [O−(M$_X$−M$_{II}$)−O]$^{2+}$ = [O−(M$_X$−V$_O^{2+}$−M$_{II}$)−O] = [O−(M$_X$−M$_{II}^{2+}$)−O] (X is whatever the valence is). When both Ms are not divalent, such as trivalent In and Ga (M$_{III}$), it is tempting to describe [O−(M$_{III}$−M$_{III}$)−O]$^{2+}$ as [O−(M$_{III}$−V$_O^{2+}$−M$_{III}$)−O] and [O−(M$_{III}$−M$_{III}$)−O]$^{3+}$ as [O−(M$_{III}$−M$_{III}^{3+}$)−O]. However, when both the V$_O$ and M$_{III}$ are shallow donors, where the conduction electrons come from is not clearly identified, and [O−(M$_{III}$−M$_{III}$)−O]$^{2+}$ and [O−(M$_{III}$−M$_{III}$)−O]$^{3+}$ are essentially indistinguishable: 3[O−(M$_{III}$−M$_{III}$)−O]$^{2+}$ = 2[O−(M$_{III}$−M$_{III}$)−O]$^{3+}$+[O−(M$_{III}$−M$_{III}$)−O]$^{0}$. Thus, there is still a problem with the definition of vacancy and interstitial defects in amorphous solids, even with considering metal valency. Meta-stable perrhenate (O$_2^{2−}$) defects that are created by excited holes$^{32}$ and [O$_2^{2−}$+2H$^+_2$] defects$^{38}$ have been suggested as hole-trap centers. An excess O defect model has been previously suggested to describe electron-trap centers based on ozone-treated amorphous InGaZnO$_4$. The excess O is characterized as a weakly binding O that results in a peak at ~200°C in thermal desorption spectroscopy. Thus, the excess O can be removed using a thermal annealing process.36,39 Because electron trapping still occurs in the absence of excess O, there should be another cause of electron trapping.
In this paper, we find that undercoordinated indium (In\(^{\ast}\)) acts as an intrinsic electron-trap center in In-based amorphous oxide semiconductors. Conduction electrons are subjected to a strong conduction-electron-ion interaction near the undercoordinated In\(^{\ast}\) and trapped there, forming an In\(^{\ast}\)-M bond. The electron-trapped center is stable in the (2-\(^{\ast}\)) charge state; thus, we designate it as a negatively double-charged intrinsic (In\(^{2-}\)-M\(^{\ast}\)) center in amorphous oxide semiconductors.

**MATERIALS AND METHODS**

Amorphous InGaZnO\(_4\) is considered as a prototype In-based amorphous oxide semiconductor. For theoretical investigations, the amorphous structures are generated using a melt-and-quench molecular dynamics simulations, and the structural instability of the conduction electrons and the electronic structures are investigated using density-functional theory calculations. The projector-augmented wave pseudopotentials and the plane wave basis set with a kinetic energy cutoff of 400 eV are used. The hybrid functional of Heyd-Scuseria-Ernzerhof with a mixing parameter of 0.25 and a screening parameter of 0.2 Å\(^{-1}\) is used for the exchange-correlation energy of the electrons. A rhombohedral 112-atom supercell is adopted, and a 2 × 2 × 2 \(k\)-point mesh is used for the Brillouin zone summation. The dimer method is used to calculate the image charges in supercells using a model charge correction scheme.

**RESULTS AND DISCUSSION**

The charge density of the lowest conduction band in amorphous InGaZnO\(_4\) is shown in Figure 1a. The conduction electrons are delocalized as expected because, in amorphous InGaZnO\(_4\), the lowest conduction band states are mainly characterized by the In-5s-like atomic orbital states, and their effective overlap through the In atomic sites results in a low electron effective mass, which is the reason for the high electron mobility in amorphous InGaZnO\(_4\). Interestingly, the s-like conduction electrons in amorphous InGaZnO\(_4\) are not found to be homogeneous, but they are highly concentrated in the depicted the local atomic structure as shown in Figure 1a.

The place where the conduction electrons are highly concentrated is found to be near the undercoordinated In\(^{\ast}\) atom. In crystalline In-oxides, such as In\(_2\)O\(_3\) and crystalline InGaZnO\(_4\), the In atoms have sixfold coordination with nearby O atoms. In amorphous InGaZnO\(_4\), the coordination number of some In atoms, such as the In depicted in Figure 1a, is depleted to fivefold coordination, and the mean value of the In coordination number has been measured to be \(\sim 5.5\) (see the running coordination numbers and shaded region in Figure 1b). In Figure 1c, we plot the integrated charges in the Wigner–Seitz volume around the In atoms with a radius of 1.677 Å, as a function of the In coordination number. The In coordination number is determined by counting the number of O atoms that have a valence charge density minimum along the In–O lines higher than 0.2 ea/Å\(^3\). This criterion approximately corresponds with the number of O atoms within 2.6 Å of the central In atom. There is a tendency that the integrated charge increases as the In coordination number decreases. The In\(^{\ast}\) atom indicated by the red circle in Figure 1c is fivefold coordinated and has the highest local-integrated charge among the In atoms in the system, indicating structural instability, which will be discussed below. The local deficiency of O atoms around the In atom can accommodate the conduction electrons most likely via electrostatic attraction, which is important in ionically bonded materials. The variation in the integrated charges with the same In coordination number observed in Figure 1c can be attributed to strained In–O bonds and a variety of local-field effects in the amorphous structure. Conduction electron crowding can occur near an In atom in amorphous InGaZnO\(_4\), and the undercoordinated In atoms are more likely to be the In\(^{\ast}\) atoms, which can accommodate more conduction electrons. Conduction electron crowding does not occur at all of the undercoordinated In atoms.

![Figure 1](image-url)

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**Figure 1** (a) Charge density of the lowest conduction band in amorphous InGaZnO\(_4\). The local atomic structure is shown where the conduction electrons are crowded. The In, Ga, and Zn atoms are as indicated in the figure, and the O atoms are indicated by the small (red) atoms. The charge density isosurface is 0.001 e per supercell. (b) Running coordination numbers of In with O in crystalline (black) and amorphous (red) InGaZnO\(_4\). (c) Integrated conduction electron charge inside the Wigner–Seitz volume around the In atoms in the supercell as a function of the In coordination number. The red dot (In\(^{\ast}\)) is for the In atom shown in (a).
atoms, but at least one (In\(^{n+}\)) of the undercoordinated In atoms in the system experiences electron crowding.

The conduction electron crowding near the undercoordinated In\(^{n+}\) implies strong conduction-electron-ion interaction. We placed two electrons (2e\(^-\)) in a 112-atom supercell (1.553 \times 10^{21} \text{ cm}^{-3}) and investigated the changes in the atomic structure. The charge neutrality is satisfied by assuming a uniform background (2\(^+\)) charge. In the presence of conduction electrons, the original In\(^{n+}\) configuration (Figure 2a) is no longer stable, but a new In\(^{n-M}\) bond (in this case, an In\(^{n+}\)-Ga bond) configuration (Figure 2c) is generated. Because the conduction electrons are more concentrated near the undercoordinated In\(^{n+}\), the atomic structure near the In\(^{n+}\) atom is affected by them. We denote the original atomic configuration as the normal state (NS) and the In\(^{n-M}\) bond configuration as the electron-trapped state (In\(^{n-M}\)). The transition state (TS) between them is shown in Figure 2b.

We would like to describe the changes in the atomic structure between NS and In\(^{n-M}\). An O atom that has a tetrahedral bonding configuration with one In\(^{n+}\), one Ga and two Zn (see Figure 2a) is significantly displaced toward the midpoint between the two Zn, far away from the In\(^{n+}\) and Ga atoms (by 1.448 Å at the In\(^{n-M}\)), which results in (i) breaking two M–O (In\(^{n+}\)-O and Ga–O) bonds and (ii) formation of one new In\(^{n-M}\) (In\(^{n+}\)-Ga) bond, as shown in Figure 2c. The coordination numbers of Zn and In in the bottom left of Figure 2c are increased by 1 (from 4 to 5 for Zn and from 5 to 6 for In bonding with O) due to the structural change. This structural change reminds us of the well-known double-broken-bond DX state in semiconductors, which is formed when a donor impurity traps electrons. The In\(^{n+}\)-Ga bond (Figure 2c) is an electron trap as well, but it is intrinsic in amorphous InGaZnO\(_4\). It can be interpreted as a small polaron that is more strongly localized after forming the In\(^{n-M}\) bond.

Figure 3 shows the calculated local electronic density-of-states near the In\(^{n+}\) and Ga atoms, as the NS is transformed into the In\(^{n-M}\) structure in the (2\(^-\)) charge state. In the NS+2e\(^-\), there is a defect state inside the conduction band (indicated by In\(^{n+}\) at the top of Figure 3) that originates from the undercoordinated In\(^{n+}\) atom. The charge density shown in Figure 1a includes this defect state. As the NS+2e\(^-\) is transformed into (In\(^{n-M}\))\(^2-\), the defect level decreases; at TS\(^2-\), the defect level crosses the Fermi level near the conduction band minimum, and then, the defect state emerges inside the band gap, which is occupied by two electrons. In (In\(^{n-M}\))\(^2-\), we find a well-isolated state inside the band gap. The charge density of the (In\(^{n-M}\))\(^2-\) deep state is shown in the inset of Figure 3 and characterized by the In\(^{n+}\)-Ga σ̄ bonding and (In\(^{n+}\)/Ga)–O σ\(^-\) antibonding molecular orbitals. The deep gap state basically originates from the large double-broken-bond distortion from the conduction electrons via the strong conduction-electron-ion interaction near the undercoordinated In\(^{n+}\).

The In\(^{n+}\)-Ga σ̄ bonding and (In\(^{n+}\)/Ga)–O σ\(^-\) antibonding nature of the (In\(^{n-M}\))\(^2-\) defect state governs the structural feature of the transition between the NS+2e\(^-\) and (In\(^{n-M}\))\(^2-\). When the Fermi level is near the conduction band minimum in the NS configuration, the 2e\(^-\) conduction electrons partially occupy the In\(^{n+}\) states, which are the crowded electrons near the undercoordinated In\(^{n+}\) as shown in Figure 1. If the (In\(^{n+}\)/Ga)–O σ\(^-\) antibonding and In\(^{n+}\)-Ga σ̄ bonding levels are partially occupied (from NS+2e\(^-\) to TS\(^2-\)), the (In\(^{n+}\)/Ga)–O bonds are broken, with the distances between the (In\(^{n+}\)/Ga) and O further increased via the formation of the In\(^{n+}\)-Ga bond, and the structure is spontaneously transformed into stable (In\(^{n-M}\))\(^2-\) state. That is, the conduction electrons contribute to the formation of the (In\(^{n-M}\))\(^2-\) state through traps near the undercoordinated In\(^{n+}\) atom that shares In\(^{n+}\)-Ga σ̄ bonding and (In\(^{n+}\)/Ga)–O σ\(^-\) antibonding orbitals with nearby atoms.

The electron trap and detrap mechanisms in amorphous oxide semiconductors can, therefore, be expressed by the reaction

\[
\text{NS} + 2e^- \leftrightarrow (\text{In}^{n+} - \text{M})^{2-}. \tag{1}
\]

The calculated potential energy surfaces in the structural transition between the NS and In\(^{n-M}\) configurations are shown in Figure 4a in the neutral and (2\(^-\)) charged states. In the neutral state, only the NS is stable, whereas the (In\(^{n+}\)-M)\(^0\) is naturally unstable. In the (2\(^-\)) charge state, (In\(^{n-M}\))\(^2-\) is found to be more stable by 0.25 eV than NS+2e\(^-\). The energy barrier in the structural transition from NS+2e\(^-\) to (In\(^{n-M}\))\(^2-\) (denoted as \(\alpha\)) is 0.49 eV. The large \(\alpha\) barrier represents the energy required to fully occupy the In\(^{n+}\)-Ga σ̄ bonding and (In\(^{n+}\)/Ga)–O σ\(^-\) antibonding states in the NS configuration (as shown in Figure 3), and thus, the \(\alpha\) barrier depends on the electron carrier concentration (n). We calculate the \(\alpha\) barriers with excess electrons, that is, (3\(^-\)), (4\(^-\)), (5\(^-\)), (6\(^-\)) and (8\(^-\)), in the supercell with the...
same number of positive uniform background charges. They correspond to 1.553, 2.330, 3.106, 3.883, 4.659 and 6.212 \times 10^{21} \text{cm}^{-3}, respectively. The calculated \( \alpha \) energy barrier as a function of \( n \) is shown in Figure 4b, which is reduced with increasing \( n \). When the carrier density is \( 4.7 \times 10^{21} \text{cm}^{-3} \), the \( \alpha \) barrier is found to be zero.

The structural recovery from (In\(*-M\))\(^2\) to the NS+2e\(^-\) state can take place when the deep (In\(*-M\))\(^2\)-electronic state inside the band gap releases the two electrons. The (In\(*-M\))\(^2\)-level can be increased from (In\(*-M\))\(^2\) to TS\(^2\) in Figure 3 by thermal excitation, and when it crosses the Fermi level, the two trapped electrons are released. The recovery energy barrier (\( \beta \)) through the thermal process is calculated to be 0.74 eV, as shown in Figure 4a, which increases as the conduction electron density increases (the level of the Fermi sea is higher). The structural recovery can also occur via optical or electrical excitation of the (In\(*-M\))\(^2\)-electrons into the empty conduction bands. For the (In\(*-M\))\(^2\) \( \rightarrow \) NS+2e\(^-\)- detrapping process, the required photon energy depends on the Fermi level, and when it is at the conduction band minimum, the minimum required photon energy is estimated to be 2.1 eV.

The electron-trapping (In\(*-M\))\(^{-2}\) centers are likely to form in heavily \( n \)-doped amorphous InGaZnO\(_4\). (In\(*-M\))\(^{-2}\) acts as a donor-compensating center that reduces the electron carrier concentration. Experimentally, the carrier concentration in \( n \)-type amorphous InGaZnO\(_4\) has not surpassed \( 10^{20} \text{cm}^{-3} \) (the doping limit) by controlling oxygen partial pressure or hydrogen incorporation.\(^{54}\)

The doping limit has been measured to be much lower than the dopant concentration,\(^{51}\) implying the presence of deep electron-trapping centers in amorphous InGaZnO\(_4\).\(^{32,52}\)

The formation of (In\(*-M\))\(^2\) can also occur by optical or electrical excitation of electrons as the \( n \)-type doping in amorphous InGaZnO\(_4\). Electrical stress, positive gate bias stress (PBS) or current stress (CS), in which the \( n \)-type thin-film transistors are turned on, can be applied, and the threshold voltage has been known to be positively shifted owing to its metastability. PBS and CS generate a high concentration of carrier electrons in the amorphous InGaZnO\(_4\) channel, and via the forward reaction in Equation (1), electron trapping (In\(*-M\))\(^2\) centers can be formed. A negatively charged deep level has been hypothesized to be created in experiments, accompanied by a positive shift of the threshold voltage.\(^{20}\)

The experimentally measured thermal activation energy for electron trapping (\( E_{\text{a,trap}} \)) is in the range of 0.22–0.95 eV\(^{15-19,22}\) under PBS and 0.08–0.14 eV\(^{20}\) under CS. The \( \alpha \) energy barrier in the (In\(*-M\))\(^2\)-formation corresponds to these values, which vary depending on the carrier concentration (Figure 4b). For \( n < 10^{21} \text{cm}^{-3} \), a larger supercell is needed, which is not currently accessible, but it can be extrapolated to the \( n = 0 \) limit (\( \alpha = 5.2 \text{eV} \) and \( \beta = 0 \text{eV} \) in the neutral state as shown in Figure 4a). In the range of \( n > 10^{20} \text{cm}^{-3} \), which is typical under PBS and CS conditions, the estimated \( \alpha \) energy barriers are 0.0–1.4 eV in good agreement with the experiments (0.08–0.95 eV).\(^{15-20,22}\)

The thermal activation energy for electron detrapping (\( E_{\text{a,detrapp}} \)) (after stopping the PBS or CS) has also been measured. This value can be interpreted as the \( \beta \) energy barrier in the (In\(*-M\))\(^2\) \( \rightarrow \) NS+2e\(^-\)-transition. Without external stresses, the carrier density is typically \( n < 10^{20} \text{cm}^{-3} \) (below the doping limit) in the presence of both normal shallow donors and electron-trapping (In\(*-M\))\(^2\) centers, and the estimated \( \beta \) energy barriers are 0.0–0.7 eV in the \( n \) range. The measured values are \( E_{\text{a,detrapp}} = 0.23 \) and 0.97 eV\(^{2,19}\).

The issue that a uniform background charge with PAW formalism gives rise to an additional total energy term has been recently addressed.\(^{53}\) This term is not included in this study, and the energies obtained are only qualitative at best. For the (2–) charge state, the error is typically <0.2 eV according to reference \(^{53}\), which is smaller than the energy differences obtained in this study. Therefore, we do not need to make any qualitative changes to our conclusions. The \( \alpha \) and \( \beta \) barrier estimations shown in Figure 4b could be quantitatively affected by the additional total energy term, but their trends would be unaffected.

**CONCLUSIONS**

In conclusion, an intrinsic electron-trapping center in amorphous InGaZnO\(_4\) is identified. The conduction electrons are attracted to undercoordinated In\(^*\) and subjected to a strong electron-ion interaction. The driving force to form In\(*-M\) bonds is induced by trapped electrons. The negatively double-charged (electron-trapped) intrinsic (In\(*-M\))\(^-\) centers in amorphous InGaZnO\(_4\) have an important role in pinning the Fermi level in heavily \( n \)-doped samples, and metastable positive-shifts of the threshold voltage in thin-film transistors under PBS or CS, which generate excited electrons. To suppress the PBS and CS instabilities and enhance the \( n \)-doping limit, a reduction in the number of undercoordinated In\(^*\) in amorphous InGaZnO\(_4\) is essential.

**CONFLICT OF INTEREST**

The authors declare no conflict of interest.

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Author contributions: HHN provided the main idea and performed the calculations. YSK wrote the manuscript, performed some calculations for confirmation and made the interpretations.

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