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Atomic and electronic origin of robust off-state insulation properties in Al-rich Al, Te, glass for ovonic threshold switching applications ⊘

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ABSTRACT

Ovonic threshold switching (OTS) selectors play a critical role in suppressing the sneak-path current of three-dimensional crossbar integration circuits. Compared to conventional nonmetal-telluride OTS selectors, selectors based on Al_xTe_y glass are found to have both satisfactory on-state current and selectivity. However, it is unclear why the Al-rich Al_xTe_y glass-based OTS selectors have robust insulation properties for reducing the off-state current. This work reveals the structure–property correlations of amorphous Al_xTe_y at the atomic scale by first-principles calculations. It is found that the stoichiometric Al_2Te_3 glass tends to have a clean bandgap owing to the covalent and dative bonds formed by non-equivalent sp^3 -hybridized Al orbitals and the lone-pair electrons of Te. Unexpectedly, for Al-rich Al_xTe_y glass ($Al_{2.21}Te_{2.79}$), the Al–Al bonds formed by redundant Al-atoms have an integrated crystal orbital bond index (ICOBI) of 0.8–0.9, which is much larger than that of Al–Al bonds in pure metals (0.227), indicating they are covalent. It is the covalent Al–Al bonds that ensure the robust insulation characteristics of Al-rich Al_xTe_y glass, while the Te–Te interaction in the Al-poor Al_xTe_y glass ($Al_{1.79}Te_{3.21}$) produces midgap states, thereby reducing the insulativity. The presented atomic and electronic pictures here will provide useful theoretical insights for designing OTS selectors with improved performances.

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I. INTRODUCTION

Nonvolatile phase-change memory $(PCM)^{1-4}$ and resistive random-access memory (RRAM),⁵⁻⁹ both of which can be applied in storage-class memory $(SCM)^{10}$ and in-memory computing,^{11,12} are expected to meet the demands of data-intensive industries. Despite the crossbar integration of these nonvolatile memory units sandwiched between word lines and bit lines was developed to realize high-density storage,¹³⁻¹⁵ the essential restrictions of sneakpath current and interconnecting parasitic resistance remain.¹⁶ One promising solution is to develop a stackable selector based the on ovonic threshold switching (OTS) effect for suppressing the sneakpath current in the three-dimensional (3D) crossbar architecture.¹⁷⁻²⁰ When a threshold voltage (V_{th}) is applied, the selector is switched to the on-state, resulting in an abrupt decrease in resistance, and thus, sufficient current is provided to operate the selected memory cell. On the contrary, the selector will be in an off-state with a low voltage ($\langle V_{th} \rangle$) so as to prevent the crossbar sneak-path current. In general, an ideal OTS selector is required to have both a sufficiently high on-state current and a low off-state current, thereby exhibiting good selectivity.

Compared to the high off-state current of the insulatormetal-transition selector and the low on-state current of the conductive-bridging resistive switching memory-type selector, the OTS selector exhibits balanced device performances.²¹ In general, OTS selectors are fabricated based on amorphous chalcogenide glass.^{18,22–25} Among these materials, Te-based amorphous solids exhibit both satisfactory *I–V* characteristics and reliability.^{21,24,26} In addition to previously reported nonmetal-telluride materials, such as SiTe and SiGeAsTe, metal-telluride alloys also demonstrate satisfactory threshold voltage, on-state current, and selectivity.^{21,27} Recently, selectors based on amorphous Al_xTe_y alloy exhibit a low threshold voltage ($V_{th} \approx 0.7 \text{ V}$), high operating current (~1 mA), and high selectivity ($\sim 5.9 \times 10^3$), and these properties can be tuned by the compositional engineering of Al_xTe_y glass.²⁸ However, in Al_xTe_y glass alloys, the atomic and electronic pictures, as well as the mechanism by which the composition modulation of the insulation behavior, remain unclear. In particular, the question of why the Al-rich Al_xTe_y (within a moderate composition deviation from chemical stoichiometry Al2Te3) based OTS selector has robust insulation properties that lead to low off-state current, and good selectivity remains unanswered.²⁸ In general, an increase in metal atom content in a material tends to reduce its insulativity. However, in Al_xTe_y based OTS selectors, as the percentage of Al atoms increases to 44% (Al_{0.44}Te_{0.56}), the off-state current is 1.7×10^{-7} A, which is even lower than those of 3.3×10^{-7} A (Al_{0.42}Te_{0.58}) and 6.1×10^{-7} A (Al_{0.39}Te_{0.61}) at the same voltage of 0.35 V.²⁸ The atomic and electronic pictures behind are not readily clarified by direct experimental observations, which hinders the further design of metal-telluride based OTS materials.

In this work, the atomic structures and electronic properties of amorphous Al_xTe_y alloys with different compositions [Al₂Te₃ (Al_{0.40}Te_{0.60}), Al_{1.79}Te_{3.21} (Al_{0.36}Te_{0.64}), and Al_{2.21}Te_{2.79} (Al_{0.44}Te_{0.56})] are investigated by first-principles calculations and ab initio molecular dynamics (AIMD) based on density functional theory (DFT).²⁹ The main atomic structures, including short-range-order and middle-range-order motifs, are captured and mapped to structural characterization functions, such as the pair correlation function and the bond-angle distribution function. It is found that Al atoms tend to adopt a tetrahedral configuration suggesting sp³-hybridization characteristics in all of these models. Compared to the stoichiometric Al₂Te₃, under Al-poor condition $(Al_{1.79}Te_{3.21})$, the redundant Te produces midgap states that reduce the insulativity of Al_xTe_y. In contrast, under Al-rich condition $(Al_{2.21}Te_{2.79})$, no significant midgap states are observed in Al_xTe_y . This is, in part, due to the Te atoms supplying lone-pair electrons to form covalent bonds with a portion of redundant Al atoms through the formation of dative bonds. More importantly, further bonding analyses demonstrate that the Al-Al bonds introduced by the redundant Al atoms in the Al-rich sample are also covalent-like and do not produce midgap states. These results not only explain the experimentally observed robust insulation properties for reduced off-state current in Al-rich Al_xTe_y OTS devices,²⁸ but also provide theoretical references for optimizing and designing metaltelluride based OTS selector devices for high-density 3D memory applications.

II. COMPUTATIONAL METHODS

The Vienna *ab initio* simulation package (VASP)³⁰ was employed to perform the DFT calculations. The Perdew–Burke– Ernzerhof (PBE) functional was adopted to describe the electron exchange and correlation interactions.³¹ Three amorphous Al_xTe_y models were created by the melt-quenching technique (see Fig. S1 for the calculation details in the supplementary material).³² In order to compare our results with experimental reports (in which the material compositions were $Al_{0.39}Te_{0.61}$, $Al_{0.42}Te_{0.58}$, and $Al_{0.44}Te_{0.56}$, respectively),²⁸ the models with similar compositions were chosen, i.e., Al_2Te_3 ($Al_{0.40}Te_{0.60}$), $Al_{1.79}Te_{3.21}$ ($Al_{0.36}Te_{0.64}$),

and Al_{2.21}Te_{2.79} (Al_{0.44}Te_{0.56}). First, the Al_xTe_y models (each) composed of 240 atoms are diffused at 3000 K for 6 ps using AIMD with the canonical NVT ensemble. Then, an 18-ps simulation at 1300 K (close to the melting point of crystalline Al_2Te_3)^{33,34} was performed to establish a reasonable liquid phase for these models. The liquids were further quenched to 300 K at a cooling rate of 15.2 K/ps. Finally, a 15-ps simulation at 300 K was carried out to obtain the thermally equilibrated amorphous structures. The structure statistics were analyzed using the last 1000 frames (3 ps) in the MD at 300 K. Their electronic properties were analyzed based on the amorphous structures, which were further relaxed. The cutoff energy was 320 eV for all calculations. The k-points for electronic property evaluation and AIMD were $3 \times 3 \times 3$ and $1 \times 1 \times 1$, respectively. The crystal orbital Hamilton population (COHP)³⁵ and the integrated crystal orbital bond index (ICOBI)³⁶ were calculated using the Local Orbital Basis Suite Toward Electronic-Structure Reconstruction (LOBSTER) code.³⁷ The atomic structures were visualized using the VESTA code.³⁸ The bond-length cutoff for structure analyses was set as $1.15 \times R$, where R is the sum of the corresponding atomic covalent radius.

III. RESULTS AND DISCUSSION

In order to determine the atomic arrangement in amorphous Al_xTe_y with different compositions, the pair correlation function (PCF), the bond-angle distribution (BAD), and the coordination number (CN) distribution are calculated first. As seen the PCFs in Figs. 1(a)-1(c), all the models lose their long-range order (with \approx PCFs tending to 1 when the distance is greater than 8 Å), indicating that they are reasonable amorphous states. The Al-Te peaks (at ~2.66 Å) are dominant in all these $Al_x Te_y$ models, suggesting that the heteropolar Al-Te bond is the primary bonding type in these 8 amorphous phases. Under Al-poor (i.e., Te-rich) condition $(Al_{1.79}Te_{3.21})$, a small Te-Te peak (at ~3.0 Å) is presented [Fig. 1(b)], whereas under Al-rich (i.e., Te-poor) condition $(Al_{2.21}Te_{2.79})$, a small Al-Al peak (at ~2.57 Å) exists [Fig. 1(c)]. In general, the presence of a small Al-Al peak here demonstrates that the Al atoms are well-distributed. In other words, the homopolar "wrong" bonds³⁹ are not as favored in an amorphous Al_xTe_y with a composition similar to Al₂Te₃, which indicates that the material is not susceptible to phase separation. In contrast, Cu atoms in the amorphous $Ge_1Cu_2Te_3$ PCM material tend to gather⁴⁰⁻⁴² with a very significant Cu-Cu PCF peak. Moreover, the longer-distance Te-Te peak (at ~4.2 Å) and the Al-Al peak (at ~3.3 Å) imply that the amorphous Al_xTe_v models should have certain middle-range orders corresponding to specific motifs. It should be noted that even a small number of such homopolar bonds (Al-Al bonds or Te-Te bonds) will still have a significant impact on the midgap states and, therefore, change the electrical properties of amorphous Al_xTe_v.

According to the BADs shown in Figs. 1(d)-1(f), the dominantly atomic motifs are the Al-centered tetrahedrons with Te neighbors, because the predominant BAD peaks (i.e., the Te-Al-Te) of the three compositions are all centrally located around ~109° (see the dashed lines) and the CNs of Al are all nearly 4 [Figs. 1(g)-1(i)]. Figures 2(a)-2(d) present the typical Al-centered tetrahedral motifs in these amorphous phases. These motifs suggest that amorphous Al_xTe_y has a very different structure



FIG. 1. Structure characteristics of amorphous $AI_xTe_{j^*}$ (a)–(c) Pair correlation functions (PCFs), (d)–(f) bond-angle distributions (BADs), and (g)–(i) coordination number (CN) distributions of $AI_xTe_{j^*}$ with different compositions: AI_2Te_3 [(a), (d), and (g)], $AI_{1.79}Te_{3.21}$ [(b), (e), and (h)], and $AI_{2.21}Te_{2.79}$ [(c), (f), and (i)]. The dashed lines in (d), (e), and (f) highlight the bond angle of 109°, indicating the AI-centered tetrahedral motif.

compared to the conventional Te-based phase-change memory materials, which are dominated by octahedral-like motifs.^{32,43–45} Furthermore, we attempt to determine the middle-range orders in these amorphous Al_xTe_y models. Figure 2(e) shows the edged-shared-tetrahedra motif that may contribute to the ~3.3-Å Al–Al and ~4.2-Å Te–Te PCF peaks, as well as the ~77° Al–Te–Al bond angle in Figs. 1(a)–1(f). Figure 2(f) presents the corner-shared-tetrahedra motif, which may contribute to the ~4Å Al–Al PCF peak [see Figs. 1(a)–1(c)]. Figure 2(g) depicts the trigonal pyramidal Al-centered motif, where the Te–Al–Te angle is approximately 99°. Figure 2(h) shows the ethane-like motif in Al-rich amorphous Al_xTe_y which should be the origin of the ~2.57 Å Al–Al PCF peak and the Al–Al–Te bond angle in the Al-rich model [see Figs. 1(c) and 1(f)]. In addition, while the composition of Al_xTe_y changes the CN of Te, the CN of Al remains nearly unchanged [Figs. 1(g)–1(i)]. In stoichiometric amorphous Al₂Te₃, 66.91% of the Te atoms possess a CN of 3, while 32.38% of the Te atoms have a CN of 2. Owing to the minimal change in the Al CN, as the Al concentration increases, so does the CN of Te. As a result, the proportion of Te with a CN of 3 is increased to 75.31%. On the contrary, under Te-rich condition, the proportion of Te with a CN of 3 is decreased to 59.01%. The distribution of different bonds (Al–Al, Al–Te, and Te–Te) is summarized in Table I in the form of partial coordination numbers. In Al₂Te₃ and



FIG. 2. Atomic structures of (a) amorphous Ale_2Te_3 and (b) its Al-centered tetrahedron motifs highlighted. (c) Schematic of the molecular orbitals of Al–Te in (d) the Al-centered tetrahedral motif. Other typical motifs in all these Al_xTe_y models are also presented: (e) an edge-shared tetrahedral motif, (f) a corner-shared tetrahedral motif, and (g) a trigonal pyramidal Al-centered motif. (h) In the Al-rich model, an ethane-like motif exists.

Te-rich Al_{1.79}Te_{3.21}, the averaged partial CNs of Al coordinated by Al atoms (Al_{Al}) are quite low, with values of 0.045 and 0.013, respectively. However, the value of Al_{Al} increases to 0.495 in Al-rich Al_{2.21}Te_{2.79}, indicating an increase in Al–Al bonds. This result is consistent with the Al–Al PCF peak at ~2.57 Å in Fig. 1(c), and these Al–Al bonds are shown to be bonding states [see the analysis later in Fig. 5(c)]. In Al₂Te₃ and Al-rich Al_{2.21}Te_{2.79}, the averaged partial CNs of Te coordinated by Te atoms (Te_{Te}) are also quite low, with values of 0.029 and 0.022, respectively; however, the value of Te_{Te} increases to 0.375 in Te-rich Al_{1.79}Te_{3.21}, indicating an increase in Te–Te bonds. This result is consistent with the Te–Te PCF peak at ~3.0 Å in Fig. 1(b), and these Te–Te bonds are demonstrated to be anti-bonding states [see the analysis later in Fig. 5(b)].

Next, we analyze the electronic properties of amorphous Al_xTe_y with various compositions. Figure 3 shows the partial

TABLE I. Total and partial coordination number (CN) in the amorphous AI_xTe_y models. AI_{AI} and AI_{Te} represent the averaged partial CN of AI coordinated by AI and Te atoms, respectively. AI_{tot} represents the sum of AI_{AI} and AI_{Te} ; Te_{AI} and Te_{Te} represent the averaged partial CN of Te coordinated by AI and Te atoms, respectively. Te_{tot} represents the sum of Te_{AI} and Te_{Te} .

	Al _{Al}	Al_{Te}	Te _{Al}	Te _{Te}	Al _{tot}	Te _{tot}
$\begin{array}{c} Al_{1.79}Te_{3.21} \\ Al_{2}Te_{3} \\ Al_{2.21}Te_{2.79} \end{array}$	0.013	4.018	2.244	0.375	4.031	2.619
	0.045	3.981	2.654	0.029	4.026	2.683
	0.495	3.483	2.755	0.022	3.978	2.777

density of states (PDOS) and the real-space inverse participation ratio (IPR) of the energy levels.⁴⁶ In general, a larger IPR means that the state is more localized, while a smaller IPR indicates that & the state is more delocalized. The PDOS suggests that the Te atoms mainly adopt the *p*-orbital states, while the Al atoms have $s-p \sum_{n=1}^{\infty}$ orbital hybridization states (see Fig. S2 in the supplementary material as well). The Al atoms should possess sp³-hybridization characteristics according to the Al-centered tetrahedral configuration. Similar to the conventional PCM materials, such as Ge₂Sb₂Te₅,³ amorphous (stoichiometric) Al₂Te₃ has an obvious bandgap with localized band tails [Fig. 3(a)]. In contrast, localized midgap states are significantly formed under Te-rich/Al-poor condition [Fig. 3(b)], which should result from the small amount of Te-Te wrong bonds [Fig. 1(b)]. Indeed, the projected charge density of the midgap states demonstrates that they originate from Te-Te interactions (see Fig. S3 in the supplementary material), which is consistent with the valence alternation pair (VAP) model.^{22,4} Such midgap defect states can act as the trap centers for carriers^{28,48} and, thus, enhance the electrical conductivity of Te-rich/Al-poor Al_xTe_v via the trap-center-assistant tunneling process.^{22,28,48} Note that the midgap states in OTS materials have also been called defect states because of their unique local structures and roles in electronic conductivity.^{22,49} Despite the existence of Al-Al wrong bonds [Fig. 1(c)], the Al-rich model does not introduce significant midgap states and should maintain good insulation properties for reducing the off-state current [Fig. 3(c)]. In fact, the conductive mechanism in these Al_xTe_y OTS materials is mainly attributed to the electron tunneling process via the midgap defect centers.²⁸ Therefore, the conductivity





FIG. 3. Partial density of states (PDOS) and the inverse participation ratio (IPR) of amorphous (a) Al_2Te_3 , (b) $Al_{1.79}Te_{3.21}$, and (c) $Al_{2.21}Te_{2.79}$. The purple vertical lines indicate IPRs for different electronic states.

in the off-state should be primarily determined by the content and distribution of defect states, even though the bandgaps differ slightly among the three compositions (as seen in Fig. 3). As a result, the insulation properties of Al-rich Al_xTe_y could still be preserved.

It is noted that the model of Al_xTe_y in this work is a small supercell with 240 atoms, and even a small amount of such homopolar bonds in this supercell can introduce midgap states and, therefore, change the properties of the material. For example, when a small amount of Te-Te bonds appear (see PCF in Fig. 1(b)), the midgap states will be introduced in Al_xTe_y (see PDOS in Fig. 3(b)). Clearly, the content of Al-Al bonds is greater than that of Te-Te bonds. Therefore, even a small amount of Al-Al bonds can have a considerable impact on the insulation properties of Al_xTe_v . Although some methods, such as the Boltzmann transport theory, could be used to calculate the conductivity of crystals at the present stage, 50-52 it is still a challenge to directly calculate the electrical conductivity of the disordered models with several hundreds of atoms.⁵⁰⁻⁵² Moreover, the tunneling effects under an electrical field still cannot be properly treated by calculation methods based on the Boltzmann transport theory.⁵¹ Hence, we discussed the electrical properties of $Al_x Te_y$ mainly according to its static electronic structure here. Further investigations are required to determine whether the other method⁵³ is suitable for describing the electrical conductivity of OTS materials.

To understand the composition-dependent electronic mechanism, we further analyze the bonding properties of amorphous Al_xTe_y . Figures 4(a) and 4(b) show the results of the calculated electron localization function (ELF). In general, an ELF of 0.5 indicates a delocalized metallic bond, while an ELF of 1 indicates a localized covalent bond or lone-pair electrons.^{18,54} According to Figs. 4(a) and 4(b), with a high ELF (=0.9) isosurface, amorphous \approx $Al_2 Te_3$ has many localized lone-pair electrons around Te atoms, electr which agrees with other typical Te-based amorphous materials. Localized lone-pair electrons are regarded as popular in Te-based amorphous materials.⁵⁵ Figures 4(c) and 4(d) show the chargedensity difference (CDD) in the Al-rich Al_{2.21}Te_{2.79} amorphous model. The accumulated electron clouds between two atoms suggest that the majority of their chemical bonds are covalent.⁵ In particular, the CDD slices of the Al-rich model in Fig. 4(d) clearly indicate that the Al-Al bonds are also covalent-like. To confirm the bonding properties, the crystal orbital Hamilton population (COHP) [Figs. 5(a)-5(c)] and the integrated crystal orbital bond index (ICOBI) [Figs. 5(d)-5(f)] are analyzed. In general, a value of "-COHP > 0" indicates a bonding state, whereas "-COHP < 0" indicates an antibonding state. The -COHP suggests that the states close to but below the valence band maximum (VBM) for Al-Te in Al₂Te₃, Al_{1.79}Te_{3.21}, and Al_{2.21}Te_{2.79} are mainly bonding states, which is in contrast to the antibonding states of other OTS materials, such as SiTe and GeTe.¹⁸ The states close to but below the VBM for Te-Te in Al_{1.79}Te_{3.21} (the Te-rich sample) are antibonding states, whereas the states close to but below the VBM for Al-Al in Al_{2.21}Te_{2.79} (the Al-rich sample) are bonding states. Importantly, the ICOBI values of the Al-Al (0.8-0.9) and Al-Te (0.6-1) bonds demonstrate that the Al-Al bonds are covalent in the Al-rich sample, and the Al-Te bonds are covalent in all the samples.³⁶ As seen from the dashed line in Fig. 5(f), the ICOBI value of the Al-Al bond in a pure Al metal is only 0.227, which is much lower than those of Al-Al bonds (0.8-0.9) in Al-rich Al_{2.21}Te_{2.79}. This result indicates that the Al-Al bonds here are indeed covalent, which





eliminates the presence of dangling bonds and delocalized metallic bonds. Hence, the covalent Al–Al bonds in the Al-rich sample should ensure its insulation properties, which explains the low off-state current and the good selectivity observed in Al-rich Al_xTe_y based selectors.²⁸ The motifs in Fig. 4(b) are typical atomic structures of the Al₂Te₃ model [Fig. 4(a)] with lone-pair electrons. The motifs in Fig. 4(d) are the Al–Te, Al–Al, and Al–Al–Al bonds in amorphous $Al_{2.21}Te_{2.79}$ [Fig. 4(c)]. The distributions of these bonds are also included in the pair correlation functions (PCFs) and the bond-angle distributions (BADs) in Fig. 1, as well as the CNs in Table I.

Therefore, we can deduce an electronic picture of the bonding properties in amorphous Al_xTe_y alloys, as follows: (1) Under stoichiometric condition, Al atoms tend to adopt nonequivalent sp^3 hybridization,⁴⁰ while Te atoms can supply lone-pair electrons to form dative bonds with Al atoms [see Fig. 2(c)]; therefore, midgap states are not produced.⁵⁵ (2). Under Al–poor/Te–rich condition, the midgap states are produced by the Te–Te anti-bonding states [see Figs. 3(b) and 5(b)], while the edge states near VBM should be contributed by the non-bonding lone-pair electrons. The abundant lone-pair electrons provided by Te-Te bonds tend to create valence alternation pairs (VAPs) where electrons transfer from overcoordinated Te to under-coordinated $Te^{49,57,58}$ and, thus, produce the localized charged defect states in the bandgap. Indeed, we found that the midgap states in Fig. 3(b) are located around the over-coordinated Te atoms (see Fig. S3 in the supplementary material), while the localized edge states near the valence band maximum distribute around the under-coordinated Te atoms. These results agree with the feature of VAPs in chalcogenide glasses.⁵⁹ (3) Under Al-rich condition, the Al–Al bonds (formed by excessive Al atoms) are demonstrated to be covalent-like and do not result in midgap states. In brief, the robust Al-centered tetrahedral motifs, the lone-pair electron compensation from Te, and the covalent bonds between Al-Al atoms together ensure the robust off-state insulation properties of the Al-rich Al_xTe_y glass and the reported excellent selectivity of the corresponding selector device.²

It should be noted that this study mainly focused on the properties of the off-state for amorphous Al_xTe_y based OTS devices. The on-state mechanism for these disordered OTS materials is often attributed to electron transport via defect centers based on



FIG. 5. –COHP of AI–Te, AI–AI, and Te–Te in the (a) AI₂Te₃, (b) AI_{1.79}Te_{3.21}, and (c) AI_{2.21}Te_{2.79} models. (d)–(f) ICOBI (per bond) varying with the bond length for AI–Te, AI–AI, and Te–Te in the AI_xTe_y models. The dashed purple line in (f) indicates the ICOBI value of the AI–AI bond (star) in the pure AI metal.

quantum-mechanical tunneling.⁴⁸ When a high voltage is applied to the OTS device based on $Al_xTe_{y_2}$, the defect states can capture excess electrons and make them occupy shallow trap states at higher energies, where an on-state is achieved.²⁸ Therefore, a small number of midgap states can have a dominant effect on OTS properties. Nevertheless, how the microstructures in amorphous materials affect on-state performance of the device is worth investigating further in the near future.

IV. CONCLUSION

In summary, the atomic structures and electronic properties of amorphous OTS Al_xTe_y alloys are analyzed by first-principles calculations and molecular dynamics. The results demonstrate that the cation-centered (Al-centered) tetrahedral structure ensures the stability of amorphous Al_xTe_y. Strong Al-Te covalent bonds are formed by non-equivalent sp³-hybridized Al orbitals and Te lonepair electrons, resulting in a clean bandgap in the stoichiometric Al₂Te₃. Under Al-poor/Te-rich condition (Al_{1.79}Te_{3.21}), the excessive Te atoms can lead to midgap states. In contrast, the Al-Al bonds under Al-rich condition (Al2.21Te2.79) have ICOBI values of 0.8-0.9, indicating covalent bonds rather than original metallic bonds (ICOBI = 0.227) or dangling bonds and, thus, do not produce midgap states. Such a picture can explain the robust insulation properties and the low off-state current observed in the OTS selector based on Al-rich Al_xTe_y samples. Our findings here uncover the microscopic atomic structures, the bonding

mechanism, and the structure–property correlations in Al_xTe_y glass alloys, which will aid in the design of new metal-telluride based OTS selectors for 3D X-Point memory applications.

SUPPLEMENTARY MATERIAL

The melt-quenching AIMD process for obtaining amorphous Al_xTe_y models, the partial density of states (PDOS) of Al atoms in amorphous Al_xTe_y , and the partial charge densities of the midgap states that are projected to real space for $Al_{1.79}Te_{3.21}$ (the Te-rich sample) are available in the supplementary material.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Xiao-Dong Li: Data curation (lead); Investigation (lead); Writing original draft (lead). Maoan Tian: Methodology (supporting); Software (supporting). Bai-Qian Wang: Data curation (supporting); Formal analysis (supporting); Methodology (supporting). Nian-Ke Chen: Writing - original draft (equal); Writing - review & editing (lead). Xian-Bin Li: Project administration (lead); Supervision (lead); Writing - original draft (equal); Writing review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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