Electronic structures and unusually robust bandgap in an ultrahigh-mobility layered oxide semiconductor, Bi$_2$O$_2$Se

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Semiconductors are essential materials that affect our everyday life in the modern world. Two-dimensional semiconductors with high mobility and moderate bandgap are particularly attractive today because of their potential application in fast, low-power, and ultrasmall/thin electronic devices. We investigate the electronic structures of a new layered air-stable oxide semiconductor, Bi$_2$O$_2$Se, with ultrahigh mobility ($\sim 2.8 \times 10^5$ cm$^2$/V·s) and moderate bandgap ($\sim 0.8$ eV). Combining angle-resolved photoemission spectroscopy and scanning tunneling microscopy, we mapped out the complete band structures of Bi$_2$O$_2$Se with key parameters (for example, effective mass, Fermi velocity, and bandgap). The unusual spatial uniformity of the bandgap without undesired in-gap states on the sample surface with up to $\sim 50\%$ defects makes Bi$_2$O$_2$Se an ideal semiconductor for future electronic applications. In addition, the structural compatibility between Bi$_2$O$_2$Se and interesting perovskite oxides (for example, cuprate high–transition temperature superconductors and commonly used substrate material SrTiO$_3$) further makes heterostructures between Bi$_2$O$_2$Se and these oxides possible platforms for realizing novel physical phenomena, such as topological superconductivity, Josephson junction field-effect transistor, new superconducting optoelectronics, and novel lasers.

INTRODUCTION

The search for new materials with superior electronic properties is critical to the development and prosperity of the semiconductor industry. In the past decade, two-dimensional (2D) materials [for example, graphene (1–3), transition metal dichalcogenides (TMDs) (4–8), and black phosphorus (4, 9, 10)] have grown as promising candidates with great potential for future electronic applications, especially those with high carrier mobility, moderate bandgap, and ambient environment stability, and numerous 2D materials have been intensively investigated. Graphene, for example, is a robust atomically thin 2D carbon sheet with ultrahigh carrier mobility ($>10,000$ cm$^2$/V·s at room temperature) (1, 3). However, the lack of a sizeable bandgap (zero gap in monolayer graphene and very small gap for multilayer graphene) (11, 12) limits its application in field-effect devices. Few-layer TMDs, on the other hand, exhibit sizable bandgap (for example, 1.8 eV for monolayer MoS$_2$), but their application is restricted by the relatively low carrier mobility (typically less than 100 cm$^2$/V·s for MoS$_2$ thin flakes at room temperature) (5–7). Recently, few-layer black phosphorus has emerged as a good 2D semiconductor candidate, with both appreciable thickness-dependent bandgap (0.3 to 2.0 eV from bulk to monolayer) and relatively high carrier mobility ($\sim 1000$ cm$^2$/V·s at room temperature) (9), but its metastability at ambient environment (13) has hindered its potential for broad application. Therefore, the search for 2D semiconductors with excellent electronic performance and stability in the ambient environment remains urgent.

More recently, Bi$_2$O$_2$Se, an air-stable layered oxide, has emerged as a promising new semiconductor with excellent electronic properties. Its layered nature makes it ideal for fabricating electronic devices down to a few atomic layers (even monolayer), which is demonstrated in a recent study (14). The Bi$_2$O$_2$Se-based top-gated field-effect transistor device shows excellent semiconductor device properties, including high carrier mobility ($\sim 28,900$ cm$^2$/V·s at 1.9 K and $\sim 450$ cm$^2$/V·s at room temperature) and superior current on/off ratio of $>10^6$ with almost ideal subthreshold swing ($\sim 65$ mV/dec). In addition, the moderate bandgap ($\sim 0.8$ eV) of Bi$_2$O$_2$Se makes its device suitable for room temperature operation while requiring only a relatively low operation voltage [for example, compare to Si with a $1.17$-eV bandgap (15)]. These attractive properties, together with its stability in the ambient environment and easy accessibility (bulk crystal, thin film, and nanostructures are all readily accessible), make Bi$_2$O$_2$Se a promising semiconductor candidate for future ultrasmall high-performance and low-power electronic devices.

Besides its potential in electronic applications, Bi$_2$O$_2$Se is also a thermoelectric material (16), with the thermoelectric figure-of-merit $ZT$ predicted as high as $1.42$ (comparable to Bi$_2$Te$_3$, one of the best thermoelectric materials broadly used today) if an in-plane strain is applied (17). Moreover, as the Bi–O layer in Bi$_2$O$_2$Se is structurally compatible...
with many perovskite oxides that exhibit rich interesting physical phenomena (for example, ferroelectricity, magnetism, multiferroics, and high-Tc superconductivity), it is possible to fabricate hybrid structures/superlattices between Bi$_2$O$_2$Se and various perovskite oxides [for example, Bi$_2$Sr$_2$Ca$_{n-1}$Cu$_n$O$_{2+\delta}$ series high-transition temperature superconductors (HTSCs) and SrTiO$_3$] to pursue novel emergent physical phenomena in hybrid semiconductor–superconductor heterostructures, such as topological superconductivity, Josephson junction field-effect transistor, new superconducting optoelectronics, and novel lasers (18–22).

To realize the full potential of Bi$_2$O$_2$Se and explore its applications in electronic, thermoelectric, and optoelectronic devices, understanding its detailed electronic structures is essential. For this purpose, we combined angle-resolved photoemission spectroscopy (ARPES) and scanning tunneling microscopy (STM) to systematically map out the full band structure of Bi$_2$O$_2$Se with key parameters including the effective mass, Fermi velocity, and the bandgap. The bandgap from both ARPES and STM shows unusual robustness and spatial uniformity without undesired in-gap (surface or edge) states—even on the cleaved sample surface with up to ~50% of Se deficiency—making Bi$_2$O$_2$Se an ideal semiconductor for future electronic applications.

**RESULTS**

**Basic characterizations**

Bi$_2$O$_2$Se crystallizes into a body-centered tetragonal structure (I4/mmm, no. 139; a = b = 3.88 Å, c = 12.16 Å) with a repeating sequence of \(-(Bi_2O_2)_{1-}\text{Se}_1-(Bi_2O_2)_{2-}\text{Se}_2-\ldots\) layers, as illustrated in Fig. 1A. High-quality Bi$_2$O$_2$Se single bulk crystals (Fig. 1B, i) for this study were synthesized by a modified Bridgman method (see Materials and Methods for details), as verified by the x-ray diffraction (XRD) characterization (Fig. 1B, ii to iv) and the core-level photoemission spectrum (Fig. 1B, v). The Hall measurements of Bi$_2$O$_2$Se devices show a very high residual resistance ratio of 585 ($R_{xx,300K}/R_{xx,2K}$; see fig. S1A) and a superior Hall mobility of $2.8 \times 10^5$ cm$^2$/Vs at 2 K (Fig. 1C, i). We note that the metallic behavior in fig. S1A is caused by the residual carriers, which can be removed by electric gating, as demonstrated in (14). Besides, prominent Shubnikov–de Haas (SdH) quantum oscillations are also observed at low temperature (Fig. 1C, ii), indicating the long mean free path of the carriers.

In the ARPES and STM investigations, Bi$_2$O$_2$Se single crystals were cleaved inside the ultrahigh vacuum (UHV) chambers for in situ measurements. Because of the weak interaction between Bi$_2$O$_2$ and Se layers (see Fig. 1, A and D, i), the cleavage occurs on the Se plane, leaving 50% Se atoms attached to each Bi$_2$O$_2$ plane, as required by the charge neutral requirement and considering that the two Bi$_2$O$_2$ layers are symmetric on each side of the Se plane. The resulting cleaved sample surface shows an interesting intertwined weave pattern formed by Se atoms and vacancies in our STM study, which will be discussed in detail later. Surprisingly, for such a high percentage of surface defects (~50% vacancies), the ARPES measurements (Fig. 1D, ii) show a clean bandgap (indirect, ~0.8 ± 0.05 eV) between the conduction and valence bands without signatures of undesired in-gap states detrimental for the device applications (23, 24).

**Scanning tunneling spectroscopy measurements**

To confirm the nonexistence of undesired in-gap (surface or edge) states, we performed extensive STM investigations on the cleaved sample surfaces, as summarized in Fig. 2. The layered nature of Bi$_2$O$_2$Se is evident from the topography map (Fig. 2A), which shows large flat terraces with a step height of ~0.61 nm (c/2). The zoom-in STM measurements on upper and lower terraces (Fig. 1B) both illustrate the intertwined weave pattern formed by the Se vacancies that constitute ~50% of the total surface area (more details and the statistical results from larger-area STM measurements can be found in the Supplementary Materials). The Se atoms and vacancies in Fig. 2B show obvious dimerization for both Se atoms and vacancies, as we will discuss in detail later.

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**Fig. 1. General characterizations of Bi$_2$O$_2$Se single crystals.** (A) Body-center tetragonal crystal structure of Bi$_2$O$_2$Se, consisting of alternating Bi$_2$O$_2$ and Se layers. (B) (i) Optical image of a Bi$_2$O$_2$Se single crystal, showing the layered structure and shining cleaved surface. (ii to iv) XRD pattern of the (001), (100), and (010) surfaces, respectively. (v) Core-level photoemission spectrum, showing the characteristic peaks of Bi5d and Se3d levels. a.u., arbitrary units. (C) (i) Hall mobility ($\mu_{\text{Hall}}$) and carrier density ($n$) as a function of temperature in Bi$_2$O$_2$Se single crystal. (ii) SdH oscillatory part of the longitudinal magnetoresistance as a function of applied perpendicular magnetic field (the non-oscillatory background has been removed). (D) (i) Illustration of the cleavage process, leaving half Se atoms attached to each Bi$_2$O$_2$ layer (see text for more discussion). (ii) ARPES broad contour maps of conduction band (CB) minimum and valence band (VB) maximum, with the Brillouin zone (BZ) overlapped (blue frames). The indirect bandgap (~0.8 eV) is indicated.
To investigate the uniformity of the bandgap of Bi$_2$O$_2$Se, we then carried out scanning tunneling spectroscopy (STS) studies (Fig. 2, C and D). Regardless where the STS spectra were taken—from either the points far away from the step edge (for example, points P1 and P2 in Fig. 2C, i) or the points at the vicinity of the step edge (for example, points P3 and P4 in Fig. 2C, i)—the $dI/dV$ spectra all show clean bulk bandgap (~0.85 ± 0.05 eV) without any observable in-gap states, as illustrated in Fig. 2C (ii and iii). The missing in-gap states are more evident in Fig. 2D, where the $dI/dV$ spectra were taken from 16 consecutive surface positions (along a line indicated in (i)) show the uniformity of the gap size without in-gap states in (ii) and (iii), respectively. (ii) Overlapped STS spectra to demonstrate the uniform gap size. (iii) Offset STS spectra for clarity.

**ARPES measurements**

Besides the STM studies, systematic ARPES experiments were performed to reconstruct the full band structures in the complete 3D BZ. For this purpose, photon energy–dependent ARPES measurements (25,26) were carried out across a wide range (60 to 230 eV) of photon energy to cover multiple BZs along the $k_z$ direction.

As can be seen in Fig. 3A, the Fermi surface (FS) map on the $k_y$–$k_z$ plane (together with the FS map of the $k_x$–$k_y$ plane in Fig. 1F) confirms that there is only one electron pocket around the $\Gamma$ point of the BZ, which shows an ellipsoidal shape (Fig. 3B, i) with a nearly isotropic small in-plane ($k_x$–$k_y$) electron mass of $m_{\perp} = (0.14 \pm 0.02)m_e$ and an in-plane Fermi velocity as $V_{F} = (1.69 \pm 0.01) \times 10^6$ m/s. Similarly, the parameters of the hole pocket near the VBM can also be deduced from our measurements (see the Supplementary Materials for details), yielding an in-plane hole mass of $m_h = (-2.41 \pm 0.02)m_e$ along the $\Gamma$–X direction and $m_h = (-0.30 \pm 0.02)m_e$ along the X–M direction. Again, from the band structures in the full 3D BZ, there is no signature of the in-gap states at any $k_x$, $k_y$ or $k_z$ momentum [see Fig. 3, C and D, for examples of dispersions’ evolution at different $k_z$ momenta and the full band structures along the $k_x$–$k_y$ plane for $\Gamma$ ($k_z = 0$) and $Z$ ($k_z = \pi/c$) points, respectively].

**Surface topography and the robust bandgap**

After establishing the overall band structures of Bi$_2$O$_2$Se and confirming that there are no in-gap states from both ARPES and STM measurements, we now focus on the interesting half Se-covered sample surface and investigate why the massive amount of defects (~50% vacancies) does not give rise to in-gap states. For this purpose, in Fig. 4A (i to iv), we show large-scale atomic-resolution STM topography maps of the sample surface. The brightest spot in the fast Fourier transformation (FFT) of the topography map shows an obvious period of 4 × Se-Se atomic distance (Fig. 4A, ii), and both Se atoms and the vacancies dimerize and form 2 × $n$ structures (where $n$ is an integer; see Fig. 4A, iii and iv). To understand these results, we carried out ab initio calculations (using a slab model; see the Supplementary Materials for details) to estimate the formation energy of different Se-atom and vacancy configurations. Our analysis (Fig. 4A, v) shows that the chain of Se-Se dimer with one vacancy on each side (that is, $\ldots$V-Se-Se-V$\ldots$) is the most energetically favorable, thus naturally explaining the experimental observation in

![Fig. 2. Surface morphology and uniform bandgap.](http://advances.sciencemag.org/content/4/9/eaaat8355/F2.large.jpg)
Fig. 3. Complete band structure of Bi₂O₂Se. (A) (i) FS of the k_y-k_z plane showing a pocket centered at Γ with the long axis along the z direction. The blue frames (overlapped) show the k_y-k_z BZs, as illustrated in (ii). (B) Details of the electron pocket formed by the conduction band. (i) Calculated ellipsoidal FS in the 3D BZ. (ii) Stacking plot of constant energy maps showing the parabolic dispersions determined by the effective mass of the conducting electron pocket. (iii and iv) FS of the electron pocket projected onto the k_x-k_y and k_y-k_z planes, respectively, showing a nearly isotropic circular shape in (ii) and an ellipse in the k_y-k_z plane, consistent with the shape of the calculation in (i). (C) Band dispersion plots from different k_z momenta, from X-Γ-X direction (k_z = 0; bottom) to N-Z-N direction (k_z = π/c; top). No in-gap states can be seen in all plots. (D) Detailed full 3D plots of the band structures for k_z = 0 (EPhoton = 98 eV) and k_z = π/c (EPhoton = 122 eV) photons, respectively. Constant energy contours showing the band structures at different binding energies are also illustrated (on the right), indicating difference for the two different k_z values in (i) and (ii).

Fig. 4. Surface pattern formation and influence on band structure. (A) (i) STM image of cleaved Bi₂O₂Se surface illustrates a clear intertwined weave pattern. (ii) FFT of (i) shows a period of 4 × Se-Se distance along two orthogonal directions. (iii and iv) Atomically resolved STM images show Se atoms, and vacancies are dimerized along two orthogonal directions, forming periodic patterns with a period of 4 × Se-Se distance. (v) Different Se-atom and vacancy configurations (left) and the calculated formation energies (right), Se-Se dimer with one vacancy on each side has the lowest energy (right) and is thus most favorable, in agreement with the observation in (ii) to (iv). (B) (i) Monte Carlo simulation and the FFT pattern of the 50% Se–50% vacancy surface pattern using the most favorable configuration obtained from (A, v) (along both x and y directions) agree well with the measurements in (A). (ii) Statistics of the characteristic surface feature (distribution of the length of the vacancy dimers) from simulation and measurements exhibit excellent agreement. (C) (i) Illustration of STS measurements on Bi and Se atoms on the surface. (ii) Calculated STS (dI/dV) spectra of Bi and Se terminations showing no in-gap states but with different DOS on the conduction and valence bands. (iii) STS measurements show good consistency with calculation in (ii).
DISCUSSION
The robustness of the bandgap against the surface defects (~50% Se vacancies), together with the small carrier mass, moderate bandgap size, layered nature, and air stability, makes Bi₂O₂Se an ideal semiconductor for future electronic device applications, and the tunability of the electronic properties (such as the bandgap size) with thickness also makes it possible to fabricate Bi₂O₂Se-HTSC hybrid junctions.

Furthermore, besides its potential in device applications, Bi₂O₂Se also has great potential in fundamental research. For example, the Bi layer in Bi₂O₂Se forms a 2D square lattice with a Bi-Bi distance of 3.88 Å, identical to that in the Bi₇Sr₇Ca₉₋ₓCuₓO_{2n+4} series of unconventional HTSC, and Bi₂O₂Se is also lattice-matched with the commonly used substrate SrTiO₃ (cubic phase; lattice constant, 3.9 Å). This lattice matching makes it possible to fabricate Bi₂O₂Se-HTSC hybrid junctions and novel interface 2D electron gas (for example, in Bi₂O₂Se-SrTiO₃ heterostructure), thus opening the door for the study of numerous novel phenomena, such as topological superconductivity.

Sample synthesis
Bi₂O₂Se single crystals were prepared via a modified Bridgman method. The stoichiometric high-purity Bi₂O₃ powder (99.999%), Se powder (99.9999%), and Bi powder (99.999%) were weighted into evacuated quartz tube with pressure down to 10⁻⁵ Pa. The Bi₂O₂Se powder was obtained when the temperature was kept at 773 K for 6 hours. As-synthesized Bi₂O₂Se was grounded into powder, reencapsulated into an evacuated quartz tube, molten at 1223 K for 2 hours, then slowly cooled down to 1123 K for 24 hours, and finally cooled down to room temperature to form a bulk crystal, which is confirmed as tetragonal Bi₂O₂Se with the space group of I4/mmm (a = b = 3.88 Å, c = 12.16 Å, Z = 2) by XRD.

Ab initio calculations
To resolve the bulk band structure of Bi₂O₂Se, we performed the first-principles calculations about Bi₂O₂Se. The density functional theory calculations were performed by using the Vienna Ab initio Simulation Package (VASP), with core electrons represented by projector-augmented wave potential. The plane-wave basis set with an energy cutoff of 400 eV was applied. To obtain accurate band structures, the modified Becke-Johnson exchange potential was adopted for the exchange-correlation functional. A k-point grid of 35 × 35 × 13 was used for the BZ sampling. Moreover, to identify the exact electronic properties of the surface pattern, we performed first-principles calculations on the Bi₂O₂Se slab model.

Supplementary materials
Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/4/9/eaat8355/DC1
Section S1. SdH quantum oscillation and effective mass in Bi₂O₂Se bulk crystal
Section S2. Statistical result of Se-atom coverage on the cleaved Bi₂O₂Se surface
Section S3. Determination of the high-symmetry points along kz and bulk band structure of Bi₂O₂Se
Section S4. Potassium doping and the structure of electron pocket
Section S5. Fitting of the electron and hole pockets
Section S6. Calculation on the formation of surface dimer
Section S7. Monte Carlo simulation and analysis of STM image
Section S8. Density functional theory calculation on half Se coverage surface
Fig. S1. SdH quantum oscillation and effective mass in Bi₂O₂Se bulk crystal
Fig. S2. STM spectra with atomic resolution in different regions.
Fig. S3. Determination of the high-symmetry points along kz and bulk band structure of Bi₂O₂Se
Fig. S4. Potassium doping and the structure of electron pocket
Fig. S5. Fitting of the electron and hole pockets
Fig. S6. Slab model used for the calculation on the formation energies of different Se-atom and vacancy configurations.
Fig. S7. Monte Carlo simulation and analysis of STM image.
Fig. S8. Theoretical calculation of half Se coverage surface.
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