**ABSTRACT**

Two-dimensional (2D) materials with high carrier mobility and tunable magnetism are in high demand for nanoelectronics and spintronic applications. Herein, we predict a novel two-dimensional monolayer KTIO that possesses an indirect band gap of 2.25 eV (based on HSE06) and high carrier mobility (1.86 \times 10^3 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1} for electron and 2.54 \times 10^3 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1} for hole) by means of *ab initio* calculations. KTIO monolayer has a calculated cleavage energy of 0.56 J m\(^{-2}\), which suggests exfoliation of bulk material as viable means for the preparation of mono- and few-layer materials. Remarkably, the KTIO monolayer suggests tunable magnetism and half-metallicity with hole doping, which are attributed to the novel Mexican-hat-like bands and van Hove singularities in its electron structure. Furthermore, monolayer KTIO exhibits moderate optical
absorption over visible light and ultraviolet region. The band gap value and band characteristics of monolayer KTlO can be strongly manipulated by biaxial and uniaxial strains to meet the requirements of various applications. All these novel properties render monolayer KTlO a promising functional material for future nanoelectronics and spintronic applications.

**KEYWORDS:** 2D materials, monolayer KTlO, mobility, magnetism, electronic properties, hole-doping, density functional theory

**I. Introduction**

Two-dimensional (2D) materials have attracted enormous attention since the successful mechanical exfoliation of graphene in 2004.\textsuperscript{1–3} To date, the family of 2D materials is growing rapidly, including elemental monolayers (such as group-III, group-IV, group-V),\textsuperscript{4–11} MXenes,\textsuperscript{12–14} transitional metal dichalcogenides (TMDCs),\textsuperscript{15–18} metal oxides\textsuperscript{19–22} and so forth.\textsuperscript{23–25} These 2D materials exhibit extraordinary properties that have been studied in various fields, such as field-effect transistors, photovoltaic solar cells and optoelectronic devices.\textsuperscript{16,26,27} In particular, 2D materials with local magnetic moments hold great potential for spintronic applications, such as spin field effect transistors, spin light-emitting diodes and solid-state quantum information processing devices.\textsuperscript{28–32} Besides using intrinsically magnetic materials, there are thus far several approaches to induce magnetism, such as introducing adatoms, defects and edges to the system. However, these approaches face serious challenges in experiments, affected by factors such as structural disorder, uncontrollable concentration of dopants and vacancies. On
the other hand, system with a Mexican-hat-like valence band maximum (VBM) may give rise to high density of states (DOS) and an almost one-dimensional-like van Hove singularity near the VBM. In these systems, hole-doping may induce a spontaneous ferromagnetic transition, observed in GaSe, α-SnO and InP$_3$ monolayers.$^{33-35}$ Compared with conventional approaches such as transition metal element doping, the magnetism in these 2D materials with Mexican-hat-like bands is inherent and can be tuned by the doping level, through liquid electrolyte gating.$^{36,37}$ To this end, searching for new candidate 2D materials with high carrier mobility and tunable magnetism is of great interest in spintronic devices.

In this work, using first-principles calculations we report a new monolayer metal shrouded semiconductor KTIO, with high dynamic and thermal stability. In addition, KTIO shows remarkably weak interlayer interactions, which result in a relatively low cleavage energy of 0.56 J m$^{-2}$. With an indirect band gap of 2.25 eV, monolayer KTIO shows high carrier mobilities of $2.54 \times 10^3$ cm$^2$ V$^{-1}$ s$^{-1}$ for electrons and $1.86 \times 10^3$ cm$^2$ V$^{-1}$ s$^{-1}$ for holes. Most fascinatingly, it exhibits extended singularity points in the DOS near the Mexican-hat shaped VBM, as well as the half-metallicity that can be tuned by hole doping. Such gate-controlled magnetism and high mobilities for electron and hole carriers in 2D KTIO render it a particularly strong candidate for spintronic devices.

II. Computational methods

All DFT calculations were performed using the plane-wave-based Vienna $Ab$-$initio$ Simulation Package (VASP) code.$^{38,39}$ The generalized gradient approximation (GGA) within the Perdew-Burke-Ernzerhof (PBE)$^{40}$ functional form was used for the
exchange-correlation energy, and projector augmented-wave (PAW) pseudopotentials\textsuperscript{41,42} were used to replace the core electrons. The Heyd–Scuseria–Ernzerhof (HSE06) screened hybrid functional\textsuperscript{35} has been used to calculate the band structures of KTIO in order to rectify the underestimated band gaps in PBE. The plane wave energy cutoff was fixed to be 500 eV. Description of the van der Waals (vdW) interactions was corrected using the DFT-D3 approach.\textsuperscript{43} For all structural relaxations, the convergence criterion for total energy was set to $1.0 \times 10^{-6}$ eV, and structural optimization was obtained until the Hellmann-Feynman force acting on each atom was less than 0.01 eV/Å for any direction. The phonon dispersion relations were calculated with the density functional perturbation theory, using the PHONOPY code.\textsuperscript{44} \textit{Ab initio} molecular dynamics (AIMD) simulations were performed to check the thermal stability of the structures, where the NVT canonical ensemble was used.

### III. Results and discussions

As shown in Fig. 1(a), the symmetry for bulk KTIO was found to be monoclinic with the space group $C2/m$, and two KTIO layers constitute the unit cell. The optimized lattice parameters of bulk KTIO were $a = 13.10$ Å, $b = 3.66$ Å, $c = 6.44$ Å and $\beta = 108.15^\circ$, in good accordance with the reported experimental results ($a = 12.87$ Å, $b = 3.62$ Å and $c = 6.29$ Å, $\beta = 106.59^\circ$).\textsuperscript{45} The two KTIO layers were joined together by vdW interactions with a interlayer distance of 3.59 Å. The layered KTIO shows a typical buckling structure, similar to Tl$_2$O and K$_2$O,\textsuperscript{46,47} while the buckling height is 3.13 Å. Interestingly, KTIO may be regarded as a ‘mixture’ of Tl$_2$O and K$_2$O with an alternately ordered combination. In addition, bulk KTIO exhibits an indirect band gap feature,
where the value was calculated to be 1.46 eV and 2.01 eV at the PBE and HSE06 levels (Fig. 1(b)), respectively. The HSE06 gap value can be regarded as a good approximation to the true fundamental gap.

**Figure 1.** (a) Crystal structure of optimized bulk KTlO, where big purple balls, medium brown balls and small red balls represent K, Tl and O atoms, respectively. (b) Calculated electronic band structures of bulk KTlO using PBE (dash line) and HSE06 (solid line) functionals, respectively. The Fermi level is set to zero energy.

Monolayer KTlO has been obtained in our simulation by taking an atomic layer from the KTlO bulk along the [001] direction, as shown in **Fig. 2(a)**. It exhibits a rectangular configuration with optimized lattice parameters of $a = 3.80$ Å and $b = 6.30$ Å. The K-O bond lengths are 2.67 Å and 2.71 Å in monolayer KTlO, while the Tl-O bond lengths are 2.54 Å and 2.40 Å, being shorter than that of Tl$_2$O (2.57 Å). The buckling height of monolayer KTlO is 2.98 Å, slightly smaller than that of bulk. Indeed, the crystal structure of the ‘mixture’ KTlO demonstrates substantial deviations comparing with the ‘clean’ Tl$_2$O and K$_2$O.

As mentioned above, KTlO shows a typical van der Waals stacking structure, thus mechanical or liquid phase exfoliation may be possible just as for graphene and black phosphorus$^{1,10,48}$. To assess such possibility, we calculated the cleavage energy of
monolayer KTIO from a five-layer KTIO slab, serving as a model of the bulk. As shown in Fig. 2(b), the cleavage energy increases with the interlayer distance, reaching a converged value of 0.56 J m\(^{-2}\). Our estimated exfoliation energies of graphene and black phosphorus are 0.32 J m\(^{-2}\) and 0.37 J m\(^{-2}\), respectively, consistent with previous theory studies.\(^{33,49}\) Therefore, it is feasible to obtain monolayer KTIO through exfoliation from the bulk, as the cleavage energy is in the same range of common 2D materials.

**Figure 2.** (a) Top and side views of a 2 \times 2 monolayer KTIO supercell, and the corresponding first Brillouin zone with high symmetry \(k\) points. The light blue square marks a unit cell of KTIO. (b) Cleavage energy estimation for the formation of monolayer KTIO, calculated by gradually enlarging the interlayer distance between a selected surface monolayer and the remainder of a five-layer slab, resembling the exfoliation process of a monolayer from the bulk model. (c) The calculated phonon dispersion spectra of monolayer KTIO. (d) Side view of the structure snapshots from the molecules dynamic simulation of the KTIO monolayer at 500 K, as well as the variation of total energy over the 10 ps simulation time.
In addition, the phonon dispersions of monolayer KTIO (shown in Fig. 2(c)) only show a tiny imaginary phonon mode (2.8 cm$^{-1}$) near the $\Gamma$ point, which comes from systematic computational error, indicating the kinetic stability. The thermal stability is further substantiated by AIMD simulations (see Fig. 2(d)), where the monolayer KTIO structure remains intact at 500 K after a 10 ps simulation time.

After verifying the stability and the feasibility of exfoliation, we turn our attention to the electronic properties of monolayer KTIO. First, to understand the bonding characteristics, we calculated its electron localization function (ELF)$^{50-53}$ and Bader charges.$^{54-56}$ As shown in Fig. 3(a), ELF = 1 corresponds to perfect localization, ELF = 0.5 corresponds to the free electron gas and ELF = 0 means the absence of electrons. For monolayer KTIO, the electron is localized around the atoms, and nearly zero along the Tl-O and K-O bonds, indicating typical ionic bond nature of Tl-O and K-O. On the other hand, the Bader charge analysis shows that 0.807 e has been transferred from a K atom to its surrounding O atoms, while the amount of charge transferred from a Tl atom is 1.01 e. For the element Tl, it can feature two stable oxidation states Tl$^+$ and Tl$^{3+}$, such as Tl$_2$O and Tl$_2$O$_3$. Yet, in monolayer KTIO, Tl prefers to be stabilized mainly as Tl$^+$ cations, similar to that of Tl$_2$O.$^{46}$ In addition, there is also extra valence electrons held in Tl atoms, which results in high electron localization around Tl atoms, as shown in Fig. 3(a). The ionic bonding nature in KTIO also implies that the effect of spin-orbit coupling (SOC) should be minor in the KTIO system, since as the only heavy element, the Tl cation has lost most of its 6$p$ electrons. To verify this argument, we examined the band structures of monolayer KTIO with and without SOC using PBE and HSE06.
According to our calculations, the difference in band gap value is less than 0.02 eV upon switching on SOC. On the other hand, the VBM and conduction band minimum (CBM) characteristics remain the same, either with or without considering SOC. Therefore, we shall neglect the SOC effect in all forthcoming band structure calculations.

Figure 3. (a) Calculated electron localization function (ELF) of monolayer KTiO, where the blue line in the upper part denotes the location for the 2D cross section demonstration of the ELF (in the lower part). (b) Band structures of monolayer KTiO calculated using PBE (red solid line) and HSE06 (blue dotted line) functionals, without considering SOC. (c) Partial DOS of the KTiO monolayer calculated using the PBE functional. The inset highlights the DOS near the Fermi level. (d) Spatial distribution of the wave-functions corresponding to the VBM and CBM of monolayer KTiO.

As shown in Fig. 3(b), monolayer KTiO exhibits an indirect band gap, calculated to be 1.47 eV and 2.25 eV according to PBE and HSE06 functionals. The CBM is located at the Y-point while the VBM lies along the Y-M direction, closer to the M-point. Note that a Mexican-hat-like dispersion was observed in monolayer KTiO,
yielding a sharp one-dimensional-like van Hove singularity in the DOS around the Fermi level as shown in Fig. 3(c). The partial DOS analysis shows that the states near the VBM mainly consist of O-3p orbitals, with a small contribution from Tl-6s and 6p orbitals. The spatial charge distributions of VBM and CBM are plotted in Fig. 3(d). The VBM charges are mainly localized at the O atom, while the Tl atoms contribute most to the CBM states.

**Figure 4.** (a) Variation of the magnetic moment and magnetic energy (per carrier) of monolayer KTlO, upon different levels of carrier doping density. (b) Spin-resolved DOS of monolayer KTlO at a hole doping density of 2.73×10^{14} cm^{-2}. The inset shows the spin density distribution, where apparently O atoms dominate the contribution.

Large DOS near the Fermi level may lead to electronic instabilities and possible phase transitions, such as magnetism and superconductivity. In the case of monolayer KTlO, we find that electronic instability can be driven by the exchange interaction from hole doping. First, the 2D KTlO monolayer was doped with holes or electrons at various carrier concentrations. Our calculations show that it spontaneously develops a ferromagnetic ground state even for a tiny amount of hole doping. **Figure 4(a)** shows the calculated electron spin magnetic moment per carrier and the magnetic energy per carrier. Here the magnetic energy is defined as the total energy difference
between the nonmagnetic and ferromagnetic states ($E_{\text{Mag}} = E_{\text{NM}} - E_{\text{FM}}$). Monolayer KTIO becomes ferromagnetic with a magnetic moment of $0.78 \mu_B$ /carrier at a very low carrier density $0.04$ hole per cell ($1.67 \times 10^{13}$ cm$^{-2}$) considered in our calculations, i.e., with a minor spin-polarization energy (1.2 meV/carrier). Then the magnetic moment saturates at nearly $0.79 \mu_B$ /carrier upon increasing hole carrier density. The magnetic moment per carrier remains at this high value even though the spin carrier density increases up to a high carrier density of $2.73 \times 10^{14}$ cm$^{-2}$. Beyond this density, monolayer KTIO returns to a nonmagnetic state. Comparing to the magnetic moment which is nearly a constant above certain carrier density, the magnetic energy has a strong dependence on the doping level. Within GGA-PBE, the magnetic energy increases nearly monotonically to $101.6$ meV/carrier at $2.73 \times 10^{14}$ cm$^{-2}$ doping level, followed by a sharp decrease back to zero at $3.57 \times 10^{14}$ cm$^{-2}$ doping level. Compared with monolayer GaSe (3 meV/carrier), the highest magnetic energy of monolayer KTIO is ~30 times larger, suggesting its superiority for spintronics. The half-metal nature is clearly demonstrated in Fig. 4(b), where the spin down states near the Fermi level are dominated by the $3p$ orbital of oxygen atoms, with a small contribution from Tl-6s and 6p orbitals. The corresponding spin density is primarily from the O and Tl atoms along the out-of-plane direction. It is even more interesting that a structural phase transition has been observed in the hole-doping process with a critical value of $0.68$ hole per unit cell (i.e. $3.57 \times 10^{14}$ cm$^{-2}$), which is also the critical point for the ferromagnetic to non-magnetic transition. The new phase is metastable and will collapse to the original structure when the doped holes disappear, indicating a repeatable hole-induced phase
transition (details are shown in Fig. S2). On the other hand, monolayer KTIO does not induce any magnetism with electron doping, but results in a semiconductor-to-metal transition (see Fig. S3), which may be of interest for nanosensors.

In order to elucidate the variations in the electronic properties of KTIO from the bulk to few layers, we have investigated the electronic band gaps of 2D KTIO with varying number of layers. The results shown in Fig. S4 are based on PBE calculations. The band gaps show a common trend of increase upon the attenuation of the film, with 1.20 eV, 1.24 eV, 1.28 eV and 1.31 eV for five-layer, four-layer, trilayer and bilayer KTIO, respectively. Similar to the bulk counterpart, all 2D KTIO multilayers under investigation have indirect band gaps. However, differences in detailed band diagram features can be observed. For bilayer KTIO, the CBM is located at the Y-point which is consistent with that of monolayer KTIO, while the VBM is located along the Γ-X direction. For tri-, four-, and five-layer KTIO, the band features are nearly the same, with the VBM located along Γ-X direction but the CBM deviated from Y. The possible reasons for band gap reduction and the band characteristics variation are the lacking of interlayer interaction as well as structural reconstruction upon stacking atomic layers.57

We further calculated the carrier mobilities (electrons and holes) of monolayer KTIO to explore its application potential in electron devices, based on the deformation potential theory proposed by Bardeen and Shockley.58 The carrier mobility of 2D materials can be calculated by the following equation:57,59,60

$$\mu_{2D} = \frac{e\hbar^2 C\varepsilon_{2D}}{k_B T m^* (E_i')^2},$$

where $\hbar$ is the reduced Planck constant, $k_B$ is the Boltzmann constant, $m^*$ is the
effective mass in the transport direction, \( m_d \) is the average effective mass determined by \( m_d = (m_a^* m_b^*)^{1/2} \), and \( T \) is the temperature (\( T = 300 \text{ K} \)). The elastic modulus \( C_{2D} \) of the longitudinal strain in the propagation direction is derived from 
\[
(E - E_0)/S_0 = C_{2D}(\Delta l/l_0)^2/2,
\]
where \( E \) is the total energy of the 2D structure, and \( S_0 \) is the lattice area of the equilibrium supercell. The deformation potential constant \( E_i^l \) is defined as \( E_i^l = \Delta E_i / (\Delta l/l_0) \). Here \( \Delta E_i \) is the energy change of the \( i \)th band under proper cell compression and dilatation (calculated using a step of 0.5%), \( l_0 \) is the lattice constant in the transport direction and \( \Delta l \) is the deformation of \( l_0 \).

**Table 1.** Calculated effective mass \( m^* \) (unit: \( m_e \)), deformation potential constant \( |E_i^l| \) (unit: eV), elastic modulus \( C_{2D} \) (unit: N m\(^{-1}\)), carrier mobility \( \mu_{2D} \) (unit: \( 10^3 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1} \)) for monolayer KTI\( \text{O} \) along the \( a \) (\( Y-M \)) direction and \( b \) (\( Y-I \)) directions.

| Carrier type | \( m_a^* \) | \( m_b^* \) | \( |E_{ia}| \) | \( |E_{ib}| \) | \( C_{2D}^a \) | \( C_{2D}^b \) | \( \mu_{2D}^a \) | \( \mu_{2D}^b \) |
|--------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|
| Electron     | 0.740       | 1.120       | 2.00        | 0.46        | 25.97       | 26.26       | 0.20        | 2.54        |
| Hole         | 2.407       | 2.434       | 0.23        | 1.54        | 25.97       | 26.26       | 1.86        | 0.04        |

Our results are summarized in **Table 1.** The elastic moduli are 25.97 N m\(^{-1}\) and 26.26 N m\(^{-1}\) along the \( a \) and \( b \) directions, respectively. They are lower than those of Tl\( \text{O} \) (\( C_a = 42.68 \text{ N m}^{-1} \) and \( C_b = 28.57 \text{ N m}^{-1} \))\(^{46}\). The effective mass of an electron is anisotropic along the \( a \) and \( b \) directions (0.740 \( m_e \) for \( a \) and 1.120 \( m_e \) for \( b \)). For the holes, the effective masses along \( a \) (2.407 \( m_e \)) and \( b \) direction (2.434 \( m_e \)) are nearly the same, and notably higher than the electrons. The large hole effective mass is due to the flat band near the VBM. The deformation potential constant \( E_1 \) shows obvious anisotropy both for the electron (1.90 and 0.26 eV) and the hole (0.46 eV and 1.54), similar to the case of Tl\( \text{O} \).\(^{46}\) The calculated electron mobility along the \( b \) direction is \( 2.54 \times 10^3 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1} \).
$V^{-1}\text{ s}^{-1}$, which is about 13 times higher than that for the $a$ direction ($0.20 \times 10^3 \text{ cm}^2 \text{ V}^{-1}\text{ s}^{-1}$), exhibiting strong anisotropy. Even stronger anisotropy is observed in the hole mobility, where the value along $a$ ($1.86 \times 10^3 \text{ cm}^2 \text{ V}^{-1}\text{ s}^{-1}$) is 46 times the value along $b$ ($0.04 \times 10^3 \text{ cm}^2 \text{ V}^{-1}\text{ s}^{-1}$), which suggests strongly direction-dependent conductivity.

**Figure 5.** Calculated absorption coefficients of monolayer KTIO.

As the band gap value of monolayer KTIO fits the energy range of visible light, we further explored its optical properties by calculating the absorption spectra in- and out-of-plane using the HSE06 functional. The transverse dielectric function $\varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega)$ is used to describe the optical properties of materials, where $\omega$ is the photon frequency, $\varepsilon_1(\omega)$ is the real part and $\varepsilon_2(\omega)$ is the imaginary part of the dielectric function, respectively. The absorption coefficient can be evaluated according to the expression\(^1\) $\alpha(\omega) = \frac{\sqrt{2\omega}}{c} \left[ \left( \varepsilon_1^2(\omega) + \varepsilon_2^2(\omega) \right)^{1/2} - \varepsilon_1(\omega) \right]^{1/2}$. As shown in **Fig. 5**, the absorption coefficients of monolayer KTIO reach the order of $10^4 \text{ cm}^{-1}$, exhibiting optical absorption over a wide wavelength range in the visible and ultraviolet light.
region. In addition, in-plane optical absorption shows strong anisotropy along the $x$ and $y$ directions. The outstanding optical properties suggest potential applications of monolayer KTIO as efficient optical absorber materials in solar cells and optoelectronic devices.

![Figure 6](image)

**Figure 6.** Electronic band gaps of monolayer KTIO under various applied strains, calculated using the PBE functional.

Finally, we studied the impact of in-plane compressive/tensile strains on the band structure of monolayer KTIO. **Figure 6** presents the PBE band gaps of monolayer KTIO under different strains in the range of -5% to 5% (the band structures are plotted in **Fig. S6**). Interestingly, the band gaps of monolayer KTIO decrease/increase monotonously with tensile/compressive strains, when it is subject to uniaxial strains along $a$ or $b$ axes. On the other hand, under the biaxial strain, the band gap value was insensitive to the magnitude of the strain, showing variations of less than 0.03 eV. In addition, the indirect bandgap feature remains unchanged within the range of strain being considered (see **Fig. S6**). The various responses to strain in monolayer KTIO may be of interest for future applications in flexible devices.
IV. Conclusions

In summary, we propose that monolayer KTIO is a remarkable new 2D semiconductor for nanoelectronics and spintronic devices. The predicted cleavage energy of 0.56 J m$^{-2}$ indicates that exfoliation from the bulk is possible to produce monolayer KTIO. Furthermore, it possesses an indirect band gap of 2.25 eV with high carrier mobilities for electrons ($2.54 \times 10^3$ cm$^2$ V$^{-1}$ s$^{-1}$) and holes ($1.86 \times 10^3$ cm$^2$ V$^{-1}$ s$^{-1}$). In particular, we find that the 2D KTIO crystal shows electron instability in its band structure, and a non-magnetic to ferromagnetic transition can be achieved by moderate hole doping within a wide range of $1.67 \times 10^{13}$ cm$^{-2}$—$2.73 \times 10^{14}$ cm$^{-2}$. Such magnetic phase transition as well as its remarkable light absorption in the range of the visible and ultraviolet light region make it a promising candidate for spintronics and optoelectronic applications.

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Supporting information for

**KTIO: A metal shrouded 2D semiconductor with high carrier mobility and tunable magnetism**

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**Figure S1.** (a) Electronic band structures of monolayer KTIO at the PBE level, with or without considering spin-orbit coupling. (b) Electronic band structures of monolayer KTIO calculated using the screened HSE06 hybrid functional, with or without considering spin-orbit coupling.
Figure S2. Crystal structures of monolayer KTIO versus various electron/hole-doping levels: (a) \(-5.2 \times 10^{13}\) cm\(^{-2}\); (b) \(0\); (c) \(1.67 \times 10^{13}\) cm\(^{-2}\); (d) \(16.54 \times 10^{13}\) cm\(^{-2}\); (e) \(27.3 \times 10^{13}\) cm\(^{-2}\); (f) \(35.7 \times 10^{13}\) cm\(^{-2}\); (g) \(53.2 \times 10^{13}\) cm\(^{-2}\).

Figure S3. Projected density of states of monolayer KTIO versus various electron/hole-doping levels: (a) \(-5.2 \times 10^{13}\) cm\(^{-2}\); (b) \(0\); (c) \(1.67 \times 10^{13}\) cm\(^{-2}\); (d) \(16.54 \times 10^{13}\) cm\(^{-2}\); (e) \(36.83 \times 10^{13}\) cm\(^{-2}\).
Figure S4. Electronic band structures of KTlO multilayers versus the number of atomic layers, calculated using the PBE functional: (a) bilayer; (b) trilayer; (c) four-layer; (d) five-layer.

Table S1. The carrier mobility $\mu_{2D} \times 10^3 \text{ cm}^2 \text{ V}^{-1}\text{s}^{-1}$ and energy bang gap (eV) at the HSE06 level ($d/i$ represent direct/indirect band gaps) of KTlO, together with the values for Ti$_2$O and K$_2$O for comparison.

| Direction | KTlO | Ti$_2$O | K$_2$O |
|-----------|------|---------|--------|
| electron  |      |         |        |
| $x$       | 0.227| 3.342   | 8.80   |
| $y$       | 1.65 | 0.404   | 18.7   |
| hole      |      |         |        |
| $x$       | 1.37 | 4.302   | 0.005  |
| $y$       | 0.04 | 0.016   | 0.0003 |
| band gap  |      |         |        |
|           | 2.25 (i) | 1.56 (d) | ~2.4 (i) |
Figure S5. (a) Relation between total energy and the applied strain $\delta$ along the $a$ (black curve) and $b$ (red curve) directions of monolayer KTiO. The quadratic fitting of the data gives the in-plane stiffness of 2D structures. (b) The VBM/CBM shifts for monolayer KTiO with respect to the vacuum energy, as a function of the applied strain along the $a$ direction. (c) The VBM/CBM shifts for monolayer KTiO with respect to the vacuum energy, as a function of the applied strain along the $b$ direction. The linear fit of the data in (b) and (c) gives the deformation potential constants.
uniaxial strain along a

uniaxial strain along b
Figure S6. Electronic band structures of monolayer KTlO under various strain situations, calculated using the PBE functional. (a) Uniaxial strain along a-axis; (b) uniaxial strain along b-axis; (c) biaxial strain.

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