MXetronics: MXene-Enabled Electronic and Photonic Devices

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ABSTRACT: MXenes are a rapidly growing family of two-dimensional (2D) materials based on transition-metal carbides and nitrides that have shown great potential as multifunctional nanomaterials. Their unique combination of metallic conductivity, hydrophilicity, and highly charged surfaces endow them with excellent electrochemical performance. Yes, these same characteristics are equally important in electronic and optoelectronic applications of MXenes, which are henceforth termed MXetronics. MXenes are suitable for solution processing in various polar solvents, allowing the preparation of MXene thin films with controlled transparency, sheet resistance, interlayer spacing, and surface chemistry. The wide selection of various MXene structures, transition-metal compositions, and surface functional groups enables tunable work function, band gap, and hence electronic and optical properties. In this Review, we summarize the recent advances in MXetronics and discuss future possibilities for this rising class of 2D materials.

Translational metal carbides and nitrides, known as MXenes, are two-dimensional (2D) derivatives of ternary layered M_{n+1}AX (MAX) phases, where M is an early transition metal, A is group IIIA–VIA element, X is carbon and/or nitrogen, and \( n = 1–3 \). The chemical reaction between MAX phases with F-containing acidic solutions results in selective extraction of M–A bonds while M–X bonds remain, followed by intercalation-mediated exfoliation. The broken M–A bonds are subject to the spontaneous formation of surface functional groups (denoted as T\_f, mainly consist of =O, –OH, –F groups), hence the commonly used MXenes have the general chemical formula M\_n\_1X\_xT\_f. The MXene family has recently been expanded to include double-transition-metal MXene either atomically ordered structures or solid solution, and vacancy-ordered structures by using different combinations of the M elements.

\( Ti_3C_2T_x \) has been the most studied MXene since its first discovery in 2011. Coexistence of metallic conductivity and hydrophilic surface featured MXenes in a wide range of applications, including high-performance electrochemical supercapacitors, catalysis, water splitting/purification, electromagnetic interference (EMI) shielding, transparent/flexible electrodes, and gas/mechanical sensors, through a cost-effective solution process. The solution-mediated deposition techniques (such as spin/spray/dip coating, layer-by-layer, drop-casting, stamping, transfer printing, etc.) are low-energy processes that can avoid unwanted damage in the underlying layers and unwanted interfacial defects/states formed by conventional high-energy processes (evaporation, sputtering, etc.). This is a potentially important aspect in MXetronics, specifically for van der Waals metal–semiconductor junctions with tunable or vanished Schottky barriers. In addition, solution deposition processes can be scaled more economically than vacuum deposition methods.

The surface functionality is one of the most interesting features of MXenes. The functional groups are strongly coupled to their properties and allow MXenes to be integrated with various organic materials/ligands. The work functions of MXenes have been reported to cover a wide range, from 1.6 eV for Sc_2C(OH)_2 to 8.0 eV for Cr_2CO_2. Oxygen and fluorine functionalization give higher work functions, because of their high electronegativity, while hydroxyl groups give an ultralow work function, because of the strong surface dipole moment. Most MXenes are known to have metallic band structures, while a few systems are expected to be semiconductors. Some MXenes have been predicted to have magnetic properties. The diverse electronic and magnetic properties of M\_xXT\_x MXenes are summarized in Figure 1, based on theoretical predictions. The reported band-gap values are also given in Table 1 for semiconducting MXenes. It is clearly seen that these characteristics are strongly dependent on the choice of transition metal atoms, surface functional

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groups, and X element (carbon and/or nitrogen). For example, the d\(^1\) transition metals (Sc and Y) in carbide MXene are predicted to be semiconductors for any type of full surface termination, while d\(^2\) transition metals (Ti, Zr, and Hf) require divalent oxygen termination to have an open band gap.\(^{22,23}\) This implies that the number of valence electron in transition metal and the electron-accepting contribution of surface functional groups may offset the free electron density in MXenes, and the strength of hybridization between them affects their band structure. Compare to the carbide MXenes, the nitride MXenes are less promising as semiconductor materials, because of the presence of an additional valence electron in each N atom; however, they may have promising magnetic properties. Specifically, Ti\(_2\)NO\(_2\), Cr\(_2\)NO\(_2\), Mn\(_2\)NO\(_2\), Mn\(_2\)N(OH)\(_2\), and Mn\(_2\)NF\(_2\) MXenes have been predicted to be ferromagnetic half-metals,\(^{24,25}\) and the Sc\(_2\)NO\(_2\) MXene has been predicted to be a ferromagnetic spin gapless semiconductor,\(^{26}\) which are promising for spintronic applications. Most of the atomically thicker M\(_3\)X\(_2\)T\(_x\) and M\(_4\)X\(_3\)T\(_x\) MXenes have a more complex band structure, because of additional states near the Fermi level; hence, many are predicted to be metallic. Few exceptions can be found from the double transition-metal MXenes listed in Table 1.

Note that experimentally synthesized Ti\(_3\)CT\(_x\) MXene thin films show metallic behavior.\(^{30}\) The difference between theoretical prediction and experimentally obtained results can be attributed to the mixed surface groups, the presence of intercalated Li\(^+\) cations, and possible defects formed during the synthesis process. The use of Li\(^+\) cation in Ti\(_3\)CT\(_x\) MXene synthesis process drastically increases their electrical conductivity, compared to classical HF etch route.\(^{51}\) In the case of MoS\(_2\), intercalation of Li\(^+\) turns semiconducting 2H-MoS\(_2\) into a metallic 1T-MoS\(_2\) phase.\(^{52}\) The presence of Li\(^+\) cations results in spontaneous co-intercalation of water molecules between MXene nanosheets via hydrogen bonding and charge interaction. Other intercalated cations, such as Na\(^+\), K\(^+\), Rb\(^+\), Mg\(^2+\), and Ca\(^2+\), were found to modulate the Ti\(_3\)C\(_2\)T\(_x\) structural response to relative humidity and the degree of hydration;\(^{53}\) however, their effects on electronic properties are yet to be explored. Such information could have been missing, because of the highly conductive nature of Ti\(_3\)C\(_2\)T\(_x\) MXene, where the cation effect was minor. Organic intercalation methods present alternative routes to avoid atomic cations; however, this process results in significantly larger interlayer spacing that hinders inter-flake transport. Organic molecule intercalated MXenes often show semiconducting-like transport behavior, which shows a transition to metal-like behavior upon annealing.\(^{54}\) This can be explained based on thermal activation process that overcome the barrier height at the inter-flake contact points with organically intercalated MXene films.

Temperature-dependent resistance measurement of Ti\(_3\)C\(_2\)T\(_x\), Ti\(_3\)CNT\(_x\), and Mo\(_3\)TiC\(_2\)T\(_x\) by vacuum annealing with electrical biasing revealed these transitions, as shown in Figures 2a–c.\(^{54}\) Note that the formal two Ti-based MXenes were prepared via the HCl-LiF route, while the latter Mo-based MXene was prepared via the HF-TBAOH method. It can be clearly seen that Li\(^+\)-intercalated MXenes become much conductive after vacuum exposure at RT, while Mo-based MXenes without Li\(^+\) exhibit no changes. This implies that some portion of weakly bonded absorbent and water molecules

Figure 1. Summary of the theoretically predicted electronic and magnetic properties of M\(_2\)CT\(_x\) carbide MXenes and M\(_2\)NT\(_x\) nitride MXenes, where M is an early transition-metal element and T\(_x\) is a surface functional group species (O, OH, or F). The electronic band structure (metal or semiconductor) and ground-state magnetic configuration (ferromagnetic or antiferromagnetic) are shown. The diverse properties of carbide/nitride MXenes can be tuned by the choice of transition metal and surface functional group. Schematic density of state (DOS) band structures are shown on the right-hand side.
were removed under vacuum. Upon vacuum annealing, all three MXenes become more conductive, because of the removal of water, intercalant, and even surface functional groups. Ti$_3$C$_2$T$_x$ retained their metallic behavior (positive $dR/dT$), while Ti$_3$CNT$_x$ and Mo$_2$TiC$_2$T$_x$ showed semiconductor-like to metallic transition (negative to positive $dR/dT$). Figure 2d shows a schematic of the intercalant effect on electrical transport of Mo$_2$TiC$_2$T$_x$ before and after annealing. Three Mo-based MXenes (Mo$_2$CT$_x$, Mo$_2$TiC$_2$T$_x$, Mo$_2$Ti$_2$C$_3$T$_x$) synthesized via the HF-TBAOH route have shown a similar transition upon annealing under an Ar/H$_2$ ambient atmosphere (Figure 2e).\textsuperscript{55} The onset temperature of the rapid increase in electrical conductivity was found to be $\sim 220 \, ^\circ\text{C}$, which is much higher than that measured in an ambient vacuum. Intercalation of either TMA$^+$ or TBA$^+$ into Ti$_3$CNT$_x$ has been reported to result in the similar transition from metallic to semiconductor-like behavior.\textsuperscript{57} The interlayer spacing is proportional to the size of organic molecules; hence, the intercalants can engineer the resistance of MXene films.\textsuperscript{58}

Table 1. Calculated Value of the Band Gap of Semiconducting MXenes Based on Conventional (PBE) and Hybrid (HSE06) Functionals\textsuperscript{a}

| MXene termination | Band Gap [eV] | PBE | HSE06 | remark\textsuperscript{b} |
|-------------------|---------------|-----|-------|------------------------|
| Sc$_2$C O         | 1.78,\textsuperscript{27} 1.8,\textsuperscript{22} 1.84,\textsuperscript{28} 1.86,\textsuperscript{29,30} | 2.87,\textsuperscript{27} 2.9,\textsuperscript{26} 2.92,\textsuperscript{31} 3.01\textsuperscript{29} |     |     |
| Si$_2$C O         | 1.04,\textsuperscript{29,30} 1.07,\textsuperscript{31} 1.07,\textsuperscript{22,29,30} 1.05\textsuperscript{33} | 1.64,\textsuperscript{33} 1.84,\textsuperscript{26} 1.85,\textsuperscript{77} 1.88\textsuperscript{39} |     |     |
| F                 | 0.36,\textsuperscript{29,30} 0.44,\textsuperscript{27,28} 0.45,\textsuperscript{22} 0.56,\textsuperscript{30} 0.71\textsuperscript{12} | 0.71,\textsuperscript{30} 0.74,\textsuperscript{26} 0.85\textsuperscript{27} |     |     |
| Cl                | 0.88\textsuperscript{16} | 0.88\textsuperscript{16} | 1.64\textsuperscript{36} | direct band gap |
| O                 | 1.32\textsuperscript{30} | 1.32\textsuperscript{30} | 1.32\textsuperscript{30} | direct band gap |
| Ti$_3$C O         | 0.17,\textsuperscript{32} 0.24,\textsuperscript{29,34} 0.26,\textsuperscript{27} 0.32,\textsuperscript{30} 0.33\textsuperscript{15} | 0.78,\textsuperscript{30} 0.88,\textsuperscript{34} 0.92\textsuperscript{27,35} |     |     |
| Zr$_2$C O         | 0.66,\textsuperscript{32} 0.88,\textsuperscript{33} 0.95,\textsuperscript{35} 0.97,\textsuperscript{30} | 1.54,\textsuperscript{30} 1.70\textsuperscript{37} |     |     |
| Hf$_2$C O         | 0.8,\textsuperscript{52} 1.00,\textsuperscript{22,35} 1.02,\textsuperscript{37} 1.03\textsuperscript{30} | 1.66,\textsuperscript{37,35} 1.75\textsuperscript{36} |     |     |
| Cr$_2$C O         | F 0.22\textsuperscript{32} | 3.15,\textsuperscript{38} 3.49\textsuperscript{39} | AFM |     |
| OH                | 0.03\textsuperscript{32} | 1.39,\textsuperscript{38} 1.43\textsuperscript{39} | AFM |     |
| Cl                | 0.15\textsuperscript{32} | 2.56\textsuperscript{39} | AFM |     |
| Mo$_2$C O         | F 0.25,\textsuperscript{32} 0.30\textsuperscript{37} 0.86\textsuperscript{37} |     |     |
| OH                | 0.13\textsuperscript{32} |     |     |
| Cl                | 0.15\textsuperscript{32} |     |     |
| W$_2$C O         | 0.194\textsuperscript{40} | 0.472\textsuperscript{40} | TI |     |
| Mn$_2$C O         | 0.83\textsuperscript{41} |     | AFM |     |
| (Mo$_2/3$Sc$_1/3$)$_2$C O | 0.04\textsuperscript{42} | 0.58\textsuperscript{42} | piezoelectric |     |
| (Mo$_2/3$Y$_1/3$)$_2$C O | 0.04\textsuperscript{42} | 1.23\textsuperscript{42} | piezoelectric |     |
| (W$_2/3$Sc$_1/3$)$_2$C O | 0.68\textsuperscript{42} | 1.3\textsuperscript{42} | piezoelectric |     |
| (W$_2/3$Y$_1/3$)$_2$C O | 0.63\textsuperscript{42} | 1.3\textsuperscript{42} | piezoelectric |     |
| Mo$_{1.33}$C O | O$_{2/3}$F$_{1/3}$ 0.5\textsuperscript{43} |     |     |
| Hf$_2$C$_2$ O | F 1.0\textsuperscript{45} |     | AFM |     |
| Hf$_2$MoC$_2$ O | 0.24\textsuperscript{45} | 0.15\textsuperscript{44} | FM |     |
| Hf$_2$VC$_2$ F | 0.4\textsuperscript{46} | 0.9\textsuperscript{46} | multiferroic (type-II) |     |
| Mo$_2$TiC$_2$ O | 0.04,\textsuperscript{47} 0.05\textsuperscript{48} | 0.12,\textsuperscript{47} 0.13\textsuperscript{48} | TI |     |
| Mo$_2$ZrC$_2$ O | 0.07,\textsuperscript{47} 0.09\textsuperscript{48} | 0.13,\textsuperscript{47} 0.15\textsuperscript{48} | TI |     |
| Mo$_2$HfC$_2$ O | 0.15,\textsuperscript{47} 0.21\textsuperscript{48} | 0.24,\textsuperscript{47} 0.30\textsuperscript{48} | TI |     |
| W$_2$TiC$_2$ O | 0.14\textsuperscript{47} | 0.29\textsuperscript{47} | TI |     |
| W$_2$ZrC$_2$ O | 0.17\textsuperscript{47} | 0.28\textsuperscript{47} | TI |     |
| W$_2$HfC$_2$ O | 0.29\textsuperscript{47} | 0.41\textsuperscript{47} | TI |     |
| Cr$_2$TiC$_2$ F |     | 1.35\textsuperscript{58} | AFM |     |
| OH                |     | 0.85\textsuperscript{58} | AFM, direct band gap |     |
| Sc$_2$N O         | 3.23\textsuperscript{26} |     |     |     |
| Cr$_2$TiN$_2$ O | 0.5\textsuperscript{49} |     | AFM |     |
| Mn$_2$TiN$_2$ F | 1.14\textsuperscript{49} | 1.9\textsuperscript{49} | AFM |     |

\textsuperscript{a}The HSE06 approach is more accurate for band-gap prediction. MXenes with a direct band gap are marked; all others have an indirect band gap.

\textsuperscript{b}Certain MXene properties are indicated, such as antiferromagnetic (AFM), ferromagnetic (FM) in their ground state, topological insulator (TI).
modulations in work function are particularly critical to achieving MXene-contacted electronics. For example, Ti$_3$C$_2$T$_x$ MXene has been used in contact electrodes for n-type ZnO and p-type SnO channel TFTs, as well as gate electrode, achieving a high-performance CMOS inverter (see Figures 3a−d). In addition, Ti$_2$CT$_x$ MXene has recently been used as contacts in transistors with MoS$_2$, WSe$_2$, and Pentacene. The metallic conductivity of Ti$_3$C$_2$T$_x$ MXene films originates from the high free electron density in each nanosheet, and from the highly aligned structure of restacked nanosheet films providing a sufficient number of electrical paths. An ellipsometry study revealed that the real part of the dielectric constant of Ti$_3$C$_2$T$_x$ changes from positive to negative at wavelengths larger than 1130 nm, implying the onset of plasmonic responses. Intense surface plasmons have been experimentally confirmed using the high-resolution electron energy loss spectroscopy (EELS) technique. More importantly, the surface plasmon energies can be tuned by MXene’s shape, size, thickness, and surface groups that can cover a wide range of frequencies (from mid-infrared to terahertz frequencies). Figure 3e shows the visualized surface plasmon modes in the Ti$_3$C$_2$T$_x$ flake. Note that these plasmonic responses in each nanosheet remain in multilayer MXene as isolated islands, suggesting high potential in optical communication and chemical sensing applications.

Nanoindentation study on a suspended monolayer Ti$_3$C$_2$T$_x$ MXene has revealed their effective Young’s modulus of 330 GPa, which is higher than that of other solution-processed 2D materials, suggesting the potential use of MXene as multifunctional filler in composites. For example, a significantly enhanced dielectric constant was found in Ti$_3$C$_2$T$_x$ MXene/poly(vinylidene fluoride-trifluoro-ethylene-chloro-ethylenylene) (P[VDF-TrFE-CFE]) polymer composite (see Figures 3f−h). The largest enhancement was found near the percolation limit of 15.0 wt % MXene loading. Ti$_3$C$_2$T$_x$ MXene-based PVA hydrogel shows unique asymmetrical strain sensitivity with highly stretchable and self-healing ability (Figures 3i and 3j).

Given the above mentioned unique properties of MXenes and their large potential, we highlight the recent advances in MXetronics, which is a fast-emerging field of MXene.
Figure 3. (a) Schematic illustration of all-MXene contacted oxide TFTs. (b, c) Transfer characteristics of p-type SnO TFT (panel (b)) and n-type ZnO TFT (panel (c)), using Ti3C2Tx MXene films for source, drain, and gate electrodes. (d) Voltage transfer curve of a MXene-contacted CMOS inverter. [Reproduced, with permission, from ref 59. Copyright 2018, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim, Germany]. (e) High-angle annular dark-field scanning transmission electron microscopy (STEM-HAADF) image of Ti3C2Tx MXene flake with corresponding EELS fitted intensity mapping for different surface plasmonic modes. [Reproduced, with permission, from ref 60. Copyright 2018, American Chemical Society, Washington, DC.] (f) Schematic of a capacitor device using Ti3C2Tx MXene/P(VDF-TrFE-CFE) polymer composite as the dielectric. (g) TEM image showing individual Ti3C2Tx MXene flakes in the host polymer. (h) Dielectric permittivity and dielectric loss, as a function of MXene loading. The largest enhancement was found at the percolation limit of MXene loading (∼15 wt %). [Reproduced, with permission, from ref 61. Copyright 2018, American Chemical Society, Washington, DC.] (i) Photography of Ti3C2Tx MXene/PVA hydrogel composite, showing self-healing ability and stretchability. (j) Resistance changes of MXene hydrogel sensor upon repeated signature motion. [Reproduced with permission under the Creative Common Attribution NonCommercial 4.0 International License (CC BY-NC 4.0).15 Copyright 2018, American Association for the Advancement of Science (AAAS).]

research. Electronic applications of MXenes such as low-cost solution-processable electrical contact material for organic thin-film transistors (OTFTs), silicon/perovskite solar cells, charge trapping filler in polymer-based memory device, and gas sensors, as well as photonic applications including unusual plasmonic photodetection, and photoluminescent MXene quantum dots are discussed. MXenes have also been utilized as growth template for van der Waals epitaxy growth, and their hydrothermal conversion has realized robust ferroelectric single crystals with surprisingly high aspect-ratio morphology.

**ELECTRONIC APPLICATIONS OF MXENES**

Organic thin-film transistors (OTFTs) and complementary logic circuits have recently been demonstrated using Ti3C2Tx MXene as contact electrodes, with p-type Pentacene and n-type N,N-dioctyl-3,4,9,10-perylenedicarboximide (PTCDI-C8) channels (see Figure 4). A thin layer of cross-linked poly(4-vinylphenol) (cPVP) was first coated on flexible poly(ethylene 2,6-naphthalate) (PEN) substrates, followed by UV/ozone treatment through a shadow mask. The created hydrophilic/hydrophobic patterns allow selective wetting of aqueous Ti3C2Tx MXene dispersion that forms patterned MXene electrodes array after drying. This process was repeated on the top of a thicker cPVP dielectric layer for the top MXene contact electrodes. The organic channels were deposited via a thermal evaporation method through shadow masks. The work function of Ti3C2Tx was found to be 4.9 eV by Kelvin probe force microscopy (KPFM). This value is suitable for p-type Pentacene with a highest occupied molecular orbit (HOMO) level of 5.0 eV. However, pristine Ti3C2Tx was not suitable for n-type PTCDI-C8 with their lowest unoccupied molecular level (LUMO) of 4.3 eV. The work function of Ti3C2Tx MXene was effectively modulated to 4.4 eV by NH3 doping. Both p- and n-type OTFTs with proper MXene contacts show great device performance, such as maximum carrier mobility of ∼1 cm2 V−1 s−1 and on/off current ratio of ∼107. NOT, NAND, and NOR logic circuits were also demonstrated by the integration of p- and n-type OTFTs.

Ti3C2Tx MXene-contacted Si solar cells have been demonstrated with a maximum power conversion efficiency (PCE) of ∼11.5 % under AM1.5G illumination (see Figures 5a–c). Delaminated MXene solution was simply drop-casted on the groove surface of n-Si surface in n−n+p− Si solar cell. Rapid thermal annealing (RTA) treatment was adopted to improve electrical contact under an ambient Ar atmosphere for 30 s. It was also found that RTA treatment enhances the physical adhesion of MXene with the substrates. The work function of Ti3C2Tx MXene was found to be 4.28 eV, which, interestingly, was not affected by short RTA process. An ohmic
contact was formed between MXene film and the heavily doped n^-Si, along with suppressed charge carrier recombination, resulting in improved fill factor and overall PCE.

In another approach, Ti_3C_2T_x MXene was directly deposited on n^-Si substrate to form Schottky junction heterostructures for self-driven photodetectors, and a new type of solar cell with an average PCE of 10.2 % (see Figures 5d−g). The latter photovoltaic study revealed that the existence of a thin SiO_2 layer between Ti_3C_2T_x MXene and n^-Si is critical to achieving effective separation of photogenerated electron−hole pairs. A two-step chemical treatment using HCl and AuCl_3 was adopted to enhance the electrical conductivity and Schottky barrier height, respectively. The work function of MXene remained in the range of 4.8−4.9 eV. The chemical treatment using AuCl_3 also resulted in the formation of gold nanoparticles that improved the charge transfer properties of the heterostructure. Such enhancement could be affected by the plasmonic effect of gold nanoparticle, which was previously reported for surface-enhanced Raman spectroscopy application of MXene using a similar precursor, namely, HAuCl_4. The Schottky metal−semiconductor contact is also desirable in metal−semiconductor-metal planar photodetectors, as demonstrated in a recent study of gallium arsenide (GaAs) photodetector, where Ti_3C_2T_x MXene-contacted devices outperformed the standard Ti/Au-contacted device.

Recently, Ti_3C_2T_x MXene has been utilized in perovskite solar cells for work function and interface engineering, resulting in a remarkable enhancement in PCE of 26.5% over the reference device without MXene (PCE from ~15% to 19%, on average). Once a small amount of MXene is doped into the perovskite precursor solution, the work function of resulting perovskite film was effectively shifted from 4.72 eV to 4.37 eV without affecting other intrinsic properties such as band gap, the relative position of valence band to Fermi level or the film morphology. A similar work function tuning effect has been observed in solution processed TiO_2 electron transfer layer that is fine-tuned from 3.91 eV to 3.85 eV by MXene doping. The MXene-doped TiO_2 layer was subject to a thermal annealing at 480 °C in air, resulting in partial oxidation of MXene. Such work function engineering in a multilayered perovskite enabled enhanced overall solar cell performance by adjusting band alignment. In addition to the MXene doping, a thin layer of conductive MXene film was deposited to engineer the interface between perovskite absorber and TiO_2 electron donor.
transfer layer. This thin Ti₃C₂Tx MXene layer could further tune the band alignment of perovskite/TiO₂ interface by charge-transfer-induced interface dipole. Moreover, the MXene interlayer reduces the barrier height at the perovskite/TiO₂ interface and improves open-circuit voltage (\(V_{oc}\)). Each MXene adopted process contributes incremental increase in overall solar cell performances, and the best device shows a remarkable PCE of 20.14 % with a fill factor of 77.6 %, a short-circuit current density of 23.82 mA cm\(^{-2}\), and an open-circuit voltage of 1.09 V. The MXene-doped device also exhibits a reduced charge trapping efficiency, evidenced by reduced hysteresis in \(J-V\) curve and enhanced long-time exposure stability of the solar cell. The optimized MXene doping amount in the perovskite precursor solution was 0.014 mg mL\(^{-1}\).
Ti$_x$C$_2$T$_x$ MXene-based nonvolatile resistive random access memory (RRAM) has been developed in PVP composite films (Figure 6). The RRAM device was constructed in a metal-insulator-metal (MIM) sandwich structure (ITO/Ti$_x$C$_2$T$_x$@PVP/Al, from bottom to top) on the PEN substrate. The MXene-based RRAM devices show typical bipolar resistive switching behavior with excellent characteristics, such as a high ON/OFF ratio of 10$^4$ and a long retention time of $5 \times 10^4$ s. Few-atomic-layer-thick MXene allowed a low SET voltage of $0.5$ V, which is highly dependent on the MXene concentration in the composites. The larger amount of MXene results in a decrease in the spacing between the MXene sheets; hence, the formation of conductive paths becomes easier at a lower SET voltage. In contrast, RESET voltage increases due to the shorter distance between MXene, hindering the rupture of the conductive paths. The existence of surface functional groups in MXene plays a critical role for charge trapping and constructing conductive paths. Moreover, multilevel data storage could be achieved by adjusting the compliance current. In this study, MXene@PVP composite devices operate without electroforming, and the control devices made of either pure MXene or pure PVP layer does not show resistive switching behavior.

Another Ti$_x$C$_2$T$_x$ MXene-based memristor device has been reported for their use in artificial bio-synapse applications. After the electroforming process, Pt/Ti$_x$C$_2$T$_x$@Al (from bottom to top) devices show bipolar resistive switching behavior and nonvolatile memory effect. More importantly, the resistance of the device could be modulated by minimum pulsed-voltage durations of only 10 ns, which is faster than that reported in literature reports. The repeated pulsed voltage signal results in a transition from short-term plasticity to long-term plasticity, which mimics the synaptic plasticity of the human brain. The formation of oxygen vacancies was proposed to be the cause of the low resistivity state under pulsed voltages, along with the change in the oxidation state of titanium, from Ti$^{4+}$ to Ti$^{3+}$, to achieve charge balance.

Metallic Ti$_x$C$_2$T$_x$ MXene has been utilized as a gas-sensing channel for the detection of volatile organic compounds (VOCs) at the parts per billion (ppb) level (see Figures 7a–d). An ultrahigh signal-to-noise ratio (SNR) was achieved due to the low noise of metallic Ti$_x$C$_2$T$_x$ MXene and the strong signal of the fully functionalized surface of MXenes. Furthermore, high sensitivity was found especially for hydrogen-bonding gases, implying rich surface interaction between the gas molecules and the surface terminations. Metallic Ti$_x$C$_2$T$_x$ MXene shows a unique gas sensing mechanism where the resistance increases upon exposure of both oxidizing- and reducing-type gases, unlike typical semiconductor sensors involving electron donation/acceptance. The empirical limit of detection of $\sim$50 ppb for acetone, and $\sim$100 ppb for ethanol and ammonia at RT. Because of the ultra-high SNR that is 2 orders of magnitude higher than other 2D materials, the theoretically available limit of detection of Ti$_x$C$_2$T$_x$ MXene is estimated to be in the sub-ppm level for SNR of 3.

Recently, V$_2$CT$_x$ MXene-based gas sensor has been reported for their high sensitivity toward nonpolar gases (see Figures 7e–h). V$_2$CT$_x$ MXene shows effective detection of hydrogen gas, where their gas response is three times higher than that of ethanol at 100 ppm at RT. The theoretical limit of detection for hydrogen and methane was estimated to be 1.3 and 25 ppm, respectively, for SNR = 3. The sensing mechanism toward inert nonpolar gases is not clearly known yet. The gas sensing behavior of V$_2$CT$_x$ MXene is clearly different from that observed for previously studied Ti$_x$C$_2$T$_x$ MXene by the same group. This suggests the large family of MXenes as potentially promising gas sensing platform with a wide range of gas-selectivity and tunability. The roles of transition-metal species and the population of surface functional groups for...
their gas sensing mechanism are the obvious remaining questions.

Koh et al. have investigated the effect of Na⁺ cation intercalation in Ti₃C₂Tₓ MXene films and their corresponding gas sensing behavior (see Figure 8). The amount of intercalated Na⁺ cation between MXene nanosheets plays an important role in their swelling behavior and selective gas sensing performance. Specifically, Ti₃C₂Tₓ films were treated with different concentrations of NaOH solutions (0.03, 0.3, 5, and 100 mM), and their structural change of interlayer spacing was observed using in-situ XRD methods under sequential exposure of nitrogen gas and ethanol vapor. MXene film treated with 0.03 mM NaOH solution did not show noticeable changes under both ambients. At NaOH concentrations of 0.3~5 mM, MXene shows reversible shrinkage and swelling behaviors, because of dehydration and ethanol intercalation, respectively. The ethanol intercalation is thermodynamically spontaneous and driven by hydrogen bonding formation with the surface functional groups of MXene and the intercalated water molecules. No swelling was found for CO₂ gas, despite their smaller molecule size. At the higher NaOH concentration of 100 mM, similar dehydration was found without ethanol swelling. The initial dehydration upon nitrogen purging provided sites for ethanol intercalation sites; however, excessive Na⁺ ions result in stabilization of the interlayer structure. Optimum NaOH concentration was found to be 0.3 mM, where the highest swelling upon exposure of ethanol, hence, enhanced gas response and selectivity.

### PHOTONIC APPLICATIONS OF MXENES

Intense surface plasmon (SP) excitations of Ti₃C₂Tₓ MXene has first been investigated by Mauchamp et al. Unlike other 2D materials, a thickness-independent bulk plasmon energies are found from Ti₃C₂Tₓ MXene, because of their weak interlayer coupling. Later, El-Demellawi et al. successfully visualized the various SP modes using a combination of scanning transmission electron microscopy (STEM) and ultra-high resolution electron energy loss spectroscopy (EELS) for individual single-layered and multilayered Ti₃C₂Tₓ MXene. It was discovered that inherent interband transition mode and transversal SP modes are invariant across the MXene flake, while longitudinal SP modes show a variation in their SP energies, depending on the shape, size, and thickness of Ti₃C₂Tₓ flakes. Interestingly, each Ti₃C₂Tₓ nanosheets in multilayer MXene retain their own plasmonic response, demonstrating independent polarizability that is attributed to their unusual weak interlayer coupling. Furthermore, the studied Ti₃C₂Tₓ MXene flakes were vacuum annealed in situ, at temperatures up to 900 °C, to monitor the change of the surface functionalization. At temperatures above 500 °C, the desorption of fluorine was detected, which multiplies the metal-like free electron density of Ti₃C₂Tₓ flakes. As a result, the SP energy of all modes was monotonically blue-shifted.

Alshareef et al. capitalized on these useful plasmonic features and used MXenes to fabricate plasmonic photodetectors that work in the visible portion of the spectrum. Specifically, five different MXenes—Ti₃C₂Tₓ, Ti₃C₂Tₓ, Mo₂CTₓ, Nb₂CTₓ, and V₃CTₓ—were investigated for plasmonic photodetection (see Figure 9). Mo₂CTₓ exhibited the best performance among the studied MXenes in the visible spectral region. The responsivity and detectivity reached 9 A W⁻¹ and 5 × 10¹¹ Jones, respectively. The photocurrent in Mo₂CTₓ film was found to be highly dependent on their surface plasmon-assisted hot electrons. Specifically, a large photocurrent was found in the wavelength range of 400~650 nm, which overlaps with the transverse surface plasmon energy of Mo₂CTₓ MXene. The maximum I₉/I₈ ratio of ~200 was found under incident light of 660 nm and a bias of 1 V. Besides the high photodetection performance, the Mo₂CTₓ thin-film photodetector shows excellent stability under continuous illumination and repeated mechanical stress. The detailed surface plasmon energies and distribution of Mo₂CTₓ were explored and visualized by EELS.
mapping. Four distinct peaks were identified at 0.30, 0.39, 2.45, and 3.42 eV, which are assigned as longitudinal dipole, longitudinal quadrupole, transversal modes, and interband transition, respectively. Note that such multipolar surface plasmon modes have not been found in any other 2D materials, but only in MXenes. The longitudinal and transversal surface plasmon energies of MXenes are known to be further tuned by controlling the population of surface termination groups from the case of Ti$_3$C$_2$Tx MXene, which strongly relate with the free electron density of MXenes. The longitudinal dipole and quadrupole modes are also dependent on the morphology of each flake. Because of the weak interlayer interaction of MXene, each monolayer MXene flake preserves their own surface plasmon modes as isolated sheets in the multilayer film, allowing successful fabrication of broadband plasmonic photodetection.

Hydrothermal method has also been utilized to synthesize MXene quantum dots (MQDs) with photoluminescence (PL) property through a quantum confinement effect (Figures 10a–d). The first Ti$_3$C$_2$Tx MQDs have been prepared by a hydrothermal process using ammonia solution (pH ~9) at 100, 120, and 150 °C for 6 h, resulting in average particle size of 2.9, 3.7, and 6.2 nm, respectively. Most of the prepared MQDs were found to have a monolayer thickness of ~1 nm. The MQDs showed excitation-dependent PL behavior with a quantum yield of up to ~10%. The hydrothermal temperature also tunes the structure of MQDs; the pristine structure of MXene was retained up to 100 °C, a hybrid structure of Ti−C core and TiO$_2$ surface was obtained at 120 °C, and amorphous carbon dots were obtained at 150 °C. The formal two cases were found to have low cytotoxicity, and their application in multicolor cellular imaging was demonstrated. The PL intensity of all three MQDs is independent of pH, suggesting the surface passivation by −NH groups.

Recently, Ti$_3$C$_2$Tx MQDs with white PL and two-photon fluorescence (TPFL) has been synthesized via a solvothermal method (see Figures 10e–h). In this study, oleylamine (OLA) was used, instead of water, as an exfoliation medium and surface passivation agent. The synthesized MQDs with an average size of 13.1 nm show a highly preserved chemical structure of parent MXene, because of the low reaction temperature, surface passivation by −NH groups, reducing environment of OLA, and the absence of water. Under UV light excitation with the wavelength of 360 nm, MQDs dispersed in ethanol exhibits a strong PL centered at 509 nm with a wide full width at half maximum (FWHM) of 220 nm, which covers all the visible range. The photoluminescence quantum yield reaches 9.36 % established by a synergic

Figure 9. Photoresponse of Mo$_2$CT$_x$ MXene thin-film photodetectors. (a) Spectral photocurrent response under 0.7 V bias voltage. Inset is a photograph showing devices and measurement setup. (b) Linear current–voltage characteristics under different light intensities at a wavelength of 660 nm. (c) Photocurrent and responsivity as a function of light intensity. (d) Photostability under continuous illumination. (e) $I_{on}/I_{off}$ ratio of five MXene photodetectors under excitation wavelengths of 660 and 532 nm. (f) Zero-loss peak subtracted EELS spectra of a Mo$_2$CT$_x$ MXene with a truncated triangular shape. Inset figures show STEM-HAADF image and corresponding EELS fitted intensity mapping for different surface plasmonic energies: longitudinal dipole (0.3 eV), longitudinal quadrupole (0.39 eV), transversal mode (2.45 eV), and interband transition (3.42 eV). [Reproduced, with permission, from ref 75. Copyright 2019, Wiley–VCH Verlag GmbH & Co. KGaA, Weinheim, Germany.]
contribution of Ti$_3$C$_2$Tx MXene core and surface-state-linked OLA chains. More importantly, TPFL properties were found where the MQDs show a similar white luminescence spectrum under a near-infrared femtosecond pulsed layer of 800 nm.

**MXENES TEMPLATES FOR CRYSTAL GROWTH**

Ti$_3$C$_2$Tx MXene films have been used as a temporal growth template for van der Waals epitaxy growth of highly (111) oriented BaTiO$_3$ film by pulsed laser deposition (Figure 11). Because of the highly c-oriented restacking nature of MXene...
films and their hexagonal geometry in the basal plane, BaTiO$_3$ perovskite film can be grown along the (111) direction on MXene seed layer, regardless of the crystallinity of the underlying substrate. The lattice parameters of BaTiO$_3$ films
are similar to their bulk values, implying an unstrained state of van der Waals epitaxy film. The size of MXene flakes also affects the orientation of BaTiO3 films: 5-μm-sized spray-coated MXene films result in (111)-dominant growth, while 1-μm-sized dip-coated MXene films resulted in the mixed orientation of (001), (110), and (111) on amorphous quartz substrates. The surface morphology of BaTiO3 films preserves the shape of MXene flakes. BaTiO3 films grow along the (001) direction on the (111) Pt-silicon substrate, whereas the growth direction is completely converted to (111) with the MXene seed layer. More importantly, Ti3C2Tx MXene film is subject to full decomposition during deposition at 700 °C in an oxygen-rich ambient atmosphere, where the Ti atoms can incorporate into the BaTiO3 film. The resulting films grown on the MXene layer retain their dielectric constant and ferroelectric behavior.

Ferroelectric crystals of potassium niobate (KNbO3) with a high aspect ratio and uniform shape have been synthesized by using Nb2CTx MXene as precursor material in a hydrothermal conversion method (Figure 12).79 Nb2CTx MXene was subjected to simultaneous oxidation and alkalinization during the hydrothermal process in potassium hydroxide (KOH) solution with sodium dodecyl sulfate (SDS) surfactant at 190 °C for 48 h. The MXene-precipitated KNbO3 crystals exhibited robust ferroelectric properties at RT, such as a saturated polarization, a remnant polarization, and a coercive field of 21 μC cm⁻², 17 μC cm⁻², and 50 kV cm⁻¹, respectively. Under the same conduction, the use of nonlayered commercial NbC powder, instead of Nb2CTx MXene, results in cubelike KNbO3 powders with nonuniform size distribution. Without the presence of SDS surfactant, the shape of the final products is irregular. The successful synthesis of high-aspect-ratio KNbO3 ferroelectric crystal was based on the 2D layered structure of Nb2CTx MXene and shape-modulation effect of SDS. Basically, the KNbO3 inherits the 2D nature of MXene, resulting in a highly textured crystal morphology.

### SUMMARY AND OUTLOOK

MXetronics is an emerging field covering electronic, optoelectronic, and sensing applications. Despite several important developments, we believe the field is still in its infancy, and many important discoveries remain to be made. Given the large number of different MXene compositions and possible surface terminations, MXenes with variable band structures, work functions, and bandgap energies promise huge potential in many applications.

Going forward, we feel that more work is needed on new MXene compositions and surface chemistry modification, which are the key parameters to achieve a wide coverage of tunable properties. (See Figure 13.) Detailed and controllable studies are desired on the impact of such modification on the fundamental properties of MXenes and MXene-based devices. Plasma, chemical, and thermal treatment present possible approaches to modify MXene surfaces. These treatments can be used to tailor the MXene work function, which then enables its utilization to make devices such as Schottky junctions or Ohmic contacts for transistors, diodes, solar cells, and photodetectors. The surface terminations may affect other properties, such as the plasmonic behavior, thermoelectric properties, and photothermal properties of MXenes.

Simulations suggest some MXenes such as Sc2CTx, Ti3CO2, Zr2CO2, and Hf2CO2 have large semiconductor band gaps, which opens a new field of research and potential new phenomena, such as all-MXene 2D devices, field-effect devices, Schottky junctions, and memory devices. More work should be dedicated to the development of nonmetallic MXenes. The experimental realization of semiconducting MXene films remains a challenge at this stage of MXene development using solution chemistry routes. Direct growth of MXene film via physical deposition methods seems to be difficult from the viewpoint of thermodynamics; however, an epitaxially grown Ti3AlC2 MAX phase film has been reported using sputtering.86 In another approach, high-quality ultrathin α-Mo3C superconducting crystals have been synthesized via a chemical vapor deposition (CVD) process.87 Although the crystal structure of CVD-grown 2D transition-metal carbide is slightly different from that of Mo2CTx MXene, CVD may be a good option to achieve high-quality MXene films with precise control of surface functional groups for MXetronics.

Recently, some simulations have suggested Mn2C(OH)2, Mn2CF2, Ti2NO2, Cr2NO2, and Mn2NTx MXenes as ferromagnetic half-metals. These types of MXenes are potentially important in the area of spintronics research; however, experimental confirmation is required. This would also enable new functionalities and new generations of MXene devices. Double transition-metal MXenes are another important class of materials that have hardly been explored for MXetronics. The carrier concentration and type, conductivity, work function, band gaps, and surface plasmons can all be tuned by changing the combination of transition metals used in the compound. This can lead to drastic improvements in the performance of electronic and photonic devices based on MXenes.

The hydrophilic nature of Ti3C2Tx MXene allow their use in low-cost solution-processed device fabrication, because of the existence of surface functional groups and high conductive nature, unlike the complicated conversion process from graphite to reduced graphene oxide. The work function of MXene can be controlled using several different methods and should be studied in more detail. Effects of parameters such as synthesis conditions, post-synthesis heat treatment, molecular doping, and chemical treatment on work function should be thoroughly evaluated. Making MXene contact with a tunable work function can enable many types of electronic devices. The unexpected tunable multipolar surface plasmons of MXenes should also be an active area of research and further developed for novel optical communication and sensing applications. The photoluminescent MXene quantum dots have shown some initial promise in light-emitting devices, bio-imaging, and even for laser cooling through two-photon fluorescence. The use of MXene as precursors and templates for van der Waals epitaxial growth may realize many unstrained functional thin films in their closed-packed orientation, regardless of the substrate crystallinity. High-aspect-ratio single crystals with robust ferroelectric properties have been synthesized by the hydrothermal conversion of MXene. Other types of MXene-driven functional crystals are possibly obtained that should also be explored in more detail. In conclusion, when it comes to MXetronics, we believe that the best is yet to come.

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