Supplementary Information
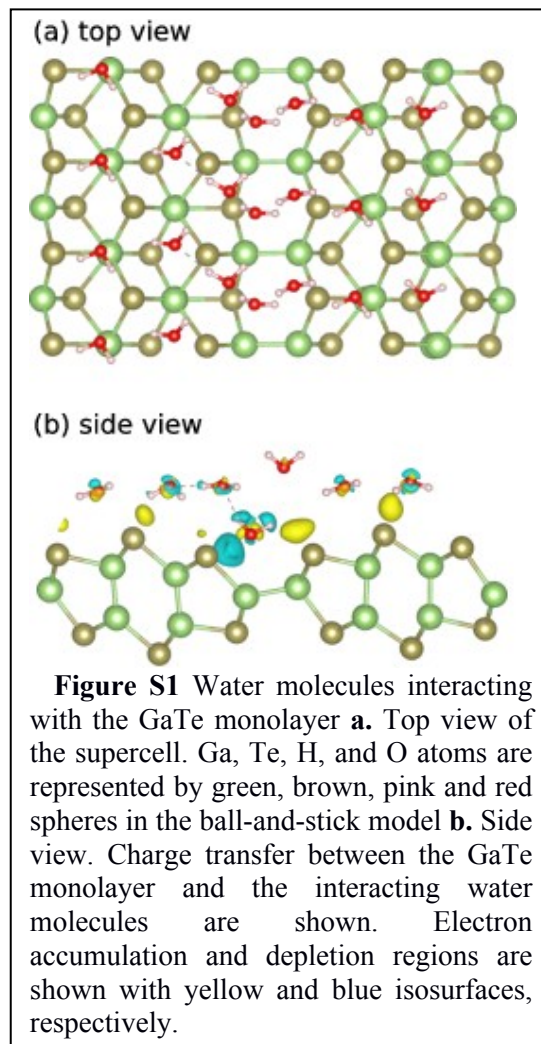
Environmental stability of 2D Anisotropic tellurium containing nanomaterials: Anisotropic to isotropic transition

Interaction between monoclinic GaTe monolayer and H₂O molecules. The interaction between monolayer gallium telluride (GaTe) with water molecules were investigated using first-principles calculations based on density functional theory (DFT). Monolayer GaTe was extracted from the bulk GaTe structure in its monoclinic phase. The electronic and geometrical parameters of the extracted monolayer were optimized to find the most favorable lattice, total energy and charge distribution in its ground state. In the first step, the optimized structure was allowed to interact with a dense coverage of water molecules as shown in **Figure S1a**. Water molecules which are in close proximity to the GaTe monolayer are attracted to the surface which results in a charge transfer from the surface to the water molecules. In this dense coverage, although water molecules are attracted to various sites on the GaTe monolayer, the strongest interaction occurs at the Te sites (in particular around where the anisotropy exists) of the GaTe monolayer which results in Te-O bonds with a binding energy of 0.28eV per water molecule. The binding energy (E_b) of a water molecule to the GaTe monolayer is calculated from the expression, $E_b=E_T[H_2O]+E_T[GaTe]-E_T[H_2O+GaTe]$, in terms of the total energies of the water molecule, GaTe monolayer and the optimized total energy of water molecule adsorbed to the GaTe monolayer. All of the total energies were calculated in the same supercell with the same convergence criterion. In our notation, $E_b >0$ indicates a binding structure.

Charge transfer characteristics. Using a similar method we also calculate the charge transfer between the surface and the water molecules, as shown in **Figure S1b**. Our results show that upon bonding, electrons (shown with yellow) transfer from the Te sites of the monolayer to the O sites of the water molecule, creating charge depleted regions on the GaTe surface (shown with blue).

Figure S1 Water molecules interacting with the GaTe monolayer **a.** Top view of the supercell. Ga, Te, H, and O atoms are represented by green, brown, pink and red spheres in the ball-and-stick model **b.** Side view. Charge transfer between the GaTe monolayer and the interacting water molecules are shown. Electron accumulation and depletion regions are shown with yellow and blue isosurfaces, respectively.

H₂O disassociation reaction at the GaTe surface. Once an H_2O molecule binds to the Te site, it is prone to disassociation into H^+ and OH^- ions with an energy barrier of 1.48 eV, as shown in the variation of total energy in **Figure S2**. As the energy of the structure increases, the water molecule first migrates towards the Ga-Ga bridge site and establishes a Ga-O bond. This breaks one of the Ga - Te bonds in the GaTe monolayer as shown in the initial structure inset of **Figure S2**. Once the total energy of the system is enough to overcome the energy barrier, water disassociates into H^+ and OH^- ions. As a result of the disassociation process, the H^+ ion binds to the Te atom while the OH^- remains bonded to the Ga atom.
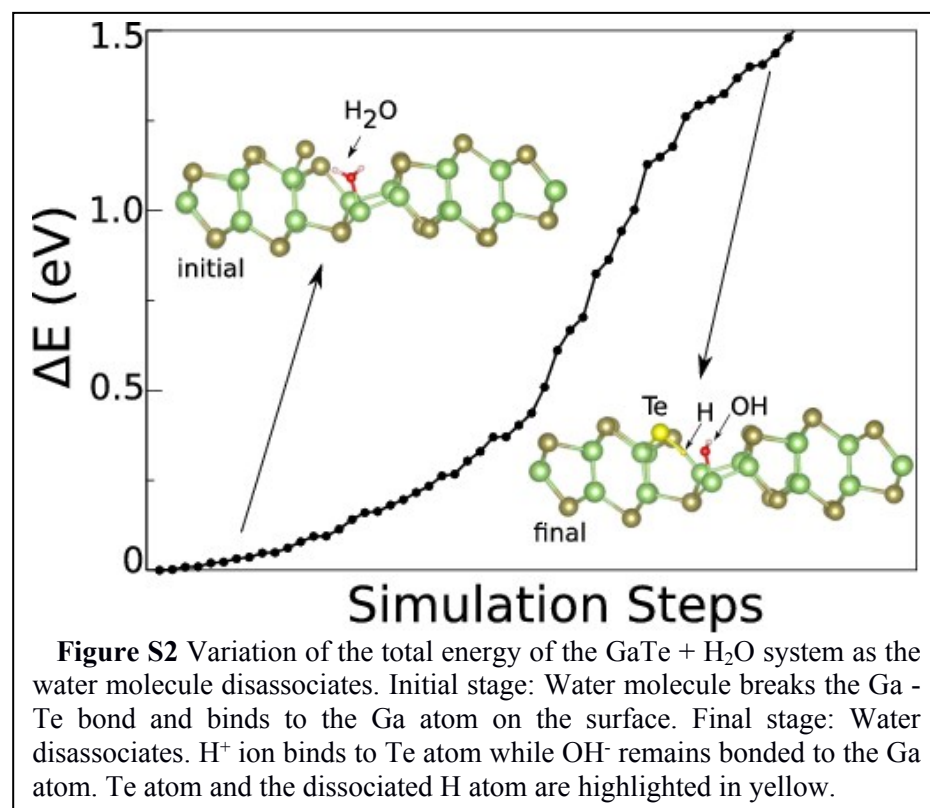
This process, which can be initiated by overcoming a small energy barrier, distorts the GaTe monolayer around the binding site as shown in the final structure in the inset of **Figure S2**.

Figure S2 Variation of the total energy of the GaTe + H_2O system as the water molecule disassociates. Initial stage: Water molecule breaks the Ga - Te bond and binds to the Ga atom on the surface. Final stage: Water disassociates. H^+ ion binds to Te atom while OH^- remains bonded to the Ga atom. Te atom and the dissociated H atom are highlighted in yellow.

Computational Method. The DFT calculations were performed within generalized gradient approximation (GGA) including van der Waals corrections[1]. We used projector-augmented wave potentials PAW [2] and the exchange-correlation potential is approximated with Perdew-Burke-Ernzerhof, PBE functional [3]. The Brillouin zone was sampled by (5x5x3) **k**-points in the Monkhorst-Pack scheme where the convergence in energy as a function of k-points was tested. The energy convergence value between two consecutive steps was chosen as 10^{-5} eV. A maximum force of 0.05 eV/A was allowed on each atom. Numerical calculations were carried out using the VASP software [4].

References

1. Grimme, S. Semiempirical GGA-Type Density Functional Constructed with a Long-Range Dispersion Correction. J. Comput. Chem. 2006, 27, 1787–1799.

2. Blöchl, P. E. Projector Augmented-Wave method. Phys. Rev. B 1994, 50, 17953–17979.

3. Perdew, J. P.; Burke, K.; Ernzerhof, M. Generalized Gradient Approximation Made Simple. Phys. Rev. Lett. 1996, 77, 3865–3868.

4. Kresse, G.; Furthmüller, J. Efficient Iterative Schemes for Ab Initio Total-energy Calculations Using a Plane-wave Basis Set. Phys. Rev. B 1996, 54, 11169–11186.