Electronic Supplementary Information (ESI) for

Two-dimensional Ga_2O_2 monolayer with tunable band gap and high hole mobility

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Computational Details

The formation energy of Ga_xO_y sheets is determined from

$$E_f = \frac{E_{Ga_xO_y} - xE_{Ga} - yE_O}{x + y}$$

where $E_{Ga_xO_y}$ is the total energy of Ga_xO_y per formula unit (f.u.) and E_{Ga} and E_O are the energies of a Ga atom in bulk metal and an O atom in O_2 ($EO = \frac{1}{2} E_{O_2}$).

The cohesive energy, E_c, is defined as

$$E_c = \frac{xE_{Ga} + yE_O - E_{Ga_xO_y}}{x + y}$$

Here $E_{Ga_xO_y}$ is the total energy of one unit cell of the Ga_2O_2 sheet or bulk Ga_2O_3, E_{Ga}/E_O represent the energy of a single Ga/O atom, respectively.

The carrier mobility was estimated by the deformation potential theory[1] using the following expression:

$$\mu = \frac{e\hbar^3 C_{2D}}{k_B T m^* m_d (E_{DP})^2}$$

Here, C_{2D}, E_{DP} and m^* represent the elastic modulus of the longitudinal strain, the deformation potential constant of the VBM for holes or CBM for electrons, and the carrier effective mass in the transport direction, respectively. The term C_{2D} is obtained from $\frac{E - E_0}{S_0} = \frac{C_{2D}}{2} (\frac{\Delta l}{l_0})^2$, where E, E_0, S_0, l_0 and Δl represent the total energy for the compressed/dilated

structure, the total energy at equilibrium, the equilibrium lattice area, the equilibrium lattice constant in the transport direction and the deformation of l_0, respectively. The term E_{DP} is given by $E_{DP} = \frac{\Delta E_i}{\Delta l / l_0}$, where ΔE_i denotes the energy change of i^{th} band under compression/dilatation from the equilibrium distance l_0 by a distance of Δl along the transport direction. The average effective mass m_d in the two directions is given by $m_d = \sqrt{m_x^* m_y^*}$. The effective mass m^* of charge carriers is calculated based on the data about the band structure (VBM for holes and CBM for electrons along x and y directions for rectangle cell) as $m^* = \hbar^2 \left[\frac{\partial^2 E(k)}{\partial k^2} \right]^{-1}$. Here x and y directions show in Fig. 4c, orthogonal to each other. The temperature T is chosen to be 300 K and ħ is reduced Planck constant. The relaxation time of the carrier is estimated by $\tau = \mu m^*/e$.

The interaction between an O_2/H_2O molecule and a monolayer is described by the adsorption energy (E_{ad}) defined as follows:

$$E_{ad} = E_{tot} - E_{Ga_2O_2} - E_{O_2/H_2O}$$

Where E_{tot}, $E_{Ga_2O_2}$, and E_{O_2/H_2O} are the energies of a Ga_2O_2 monolayer with an adsorbed O_2/H_2O molecule, the pristine monolayer, and an individual O_2/H_2O molecule, respectively. By definition, a negative E_b means an exothermic adsorption of an O_2/H_2O molecule.

The binding energy (E_b) of the Ga_2O_2 mutlilayer in different stacking configurations is calculated via $E_b = (n \times E_{monolayer} - E_{multilayer})/n$, where

$E_{monolayer}$ and $E_{multilayer}$ are the total energies of the Ga_2O_2 monolayer and multilayer, respectively. n is the number of layers.

We have also computed optical properties of the Ga_2O_2 monolayer and bilayer based on the dielectric constants, $\varepsilon(\omega)= \varepsilon_1(\omega)+i\varepsilon_2(\omega)$, where $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$ are the real and imaginary part, respectively. The absorption coefficient is given by

$$\alpha(\omega) = \sqrt{2}\omega(\sqrt{\varepsilon_1^2(\omega) + \varepsilon_2^2(\omega)} - \varepsilon_1(\omega))^{1/2}$$

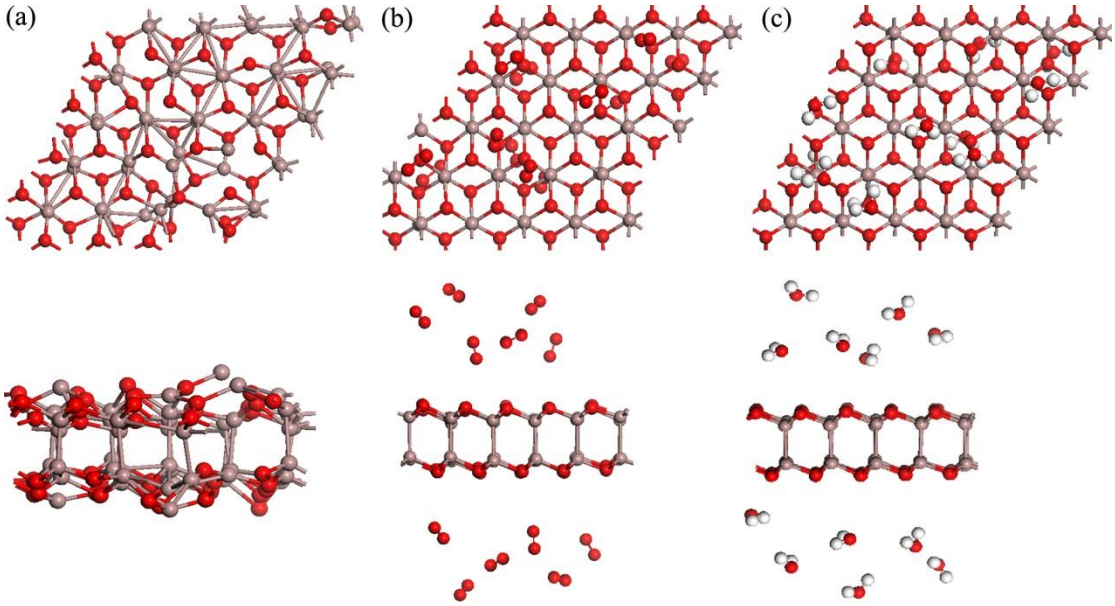

Fig. S1. Top and side views of (a) Ga_2O_2 monolayer from AIMD simulation with temperature at 1500K, and Ga_2O_2 monolayer at 300 K in the O_2 (b) and H_2O (c) atmosphere for 5 ps.

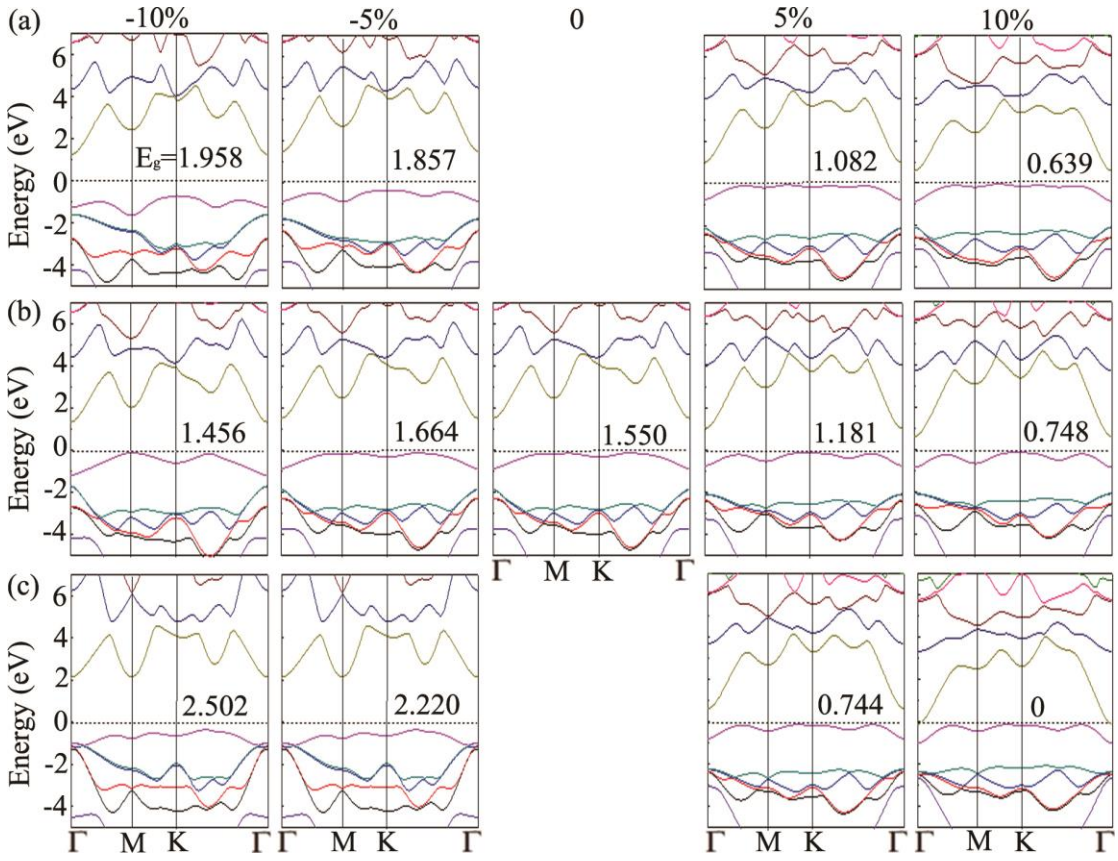

Fig. S2. Band structure changes of Ga_2O_2 sheet under a-direction (a), b-direction (b), and biaxial (c) strains at PBE level. The Fermi level is set to zero.

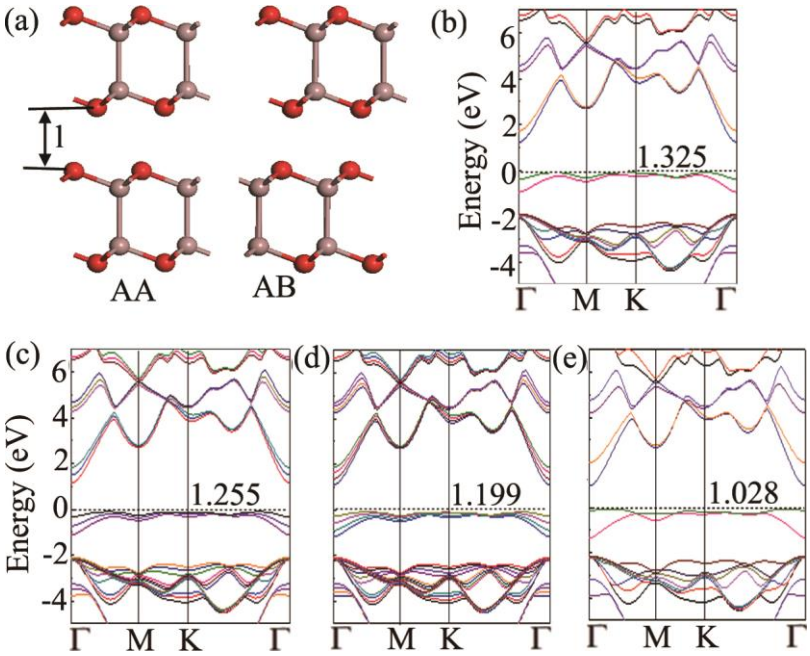

Fig. S3. AA and AB stacking configurations for Ga_2O_2 bilayer with 2×2 supercells (a). The interlayer distance l is labeled in the AA stacking. Band structure of Ga_2O_2 bilayer (b), trilayer (c), four-layer (d), and bulk (e) with AA stacking at PBE level.

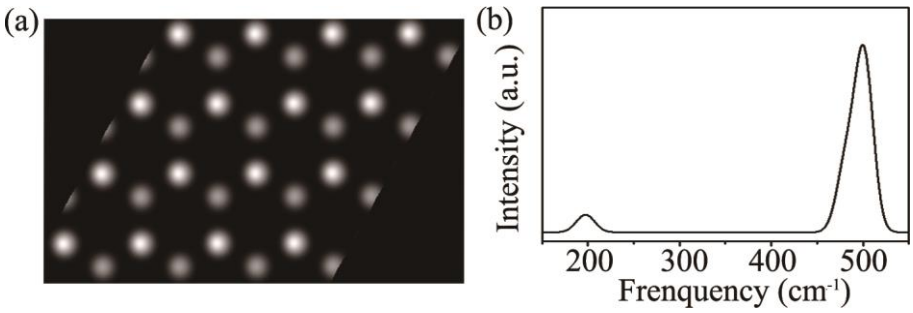

Fig. S4. Simulated STM image (a) and Raman spectrum (b) of Ga_2O_2 sheet.

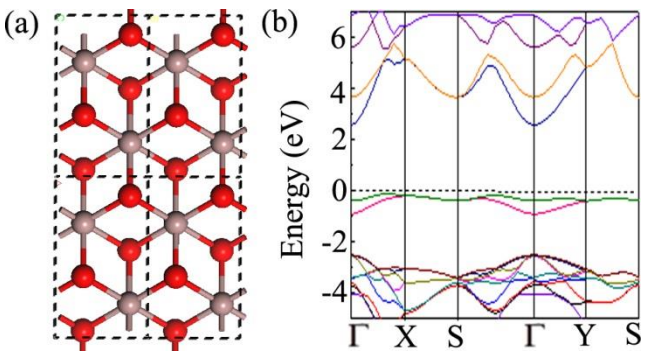

Fig. S5. (a) Atomic structure and (b) Electronic band structure of Ga_2O_2 monolayer in an orthogonal supercell.

References

(1) J. Bardeen, W. Shockley, *Phys. Rev.* 1950, **80**, 72-80.