Supplementary Information

Monolayer MoS_2 Bandgap Modulation by Dielectric Environments and Tunable Bandgap Transistors

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Methods: *GW* Calculations with Effective Environmental Dielectric Screening

Based on classical electrostatics, the Poisson equation governing an inhomogeneous dielectric system is

$$\nabla \cdot [\varepsilon^{\mathrm{M}}(\mathbf{r})\nabla V(\mathbf{r})] = -4\pi\rho(\mathbf{r}), \tag{1}$$

where $\rho(\mathbf{r})$ is the charge density, $V(\mathbf{r})$ is the electrostatic potential, and $\varepsilon^{\mathrm{M}}(\mathbf{r})$ is the spatially varying macroscopic dielectric constant. In a periodic system with the lattice vector, **R**, the Fourier transform of Eq. (1) is

$$\sum_{\mathbf{G}'} \varepsilon^{\mathrm{M}}_{\mathbf{G}-\mathbf{G}'}\mathbf{G}\cdot\mathbf{G}'V_{\mathbf{G}'} = 4\pi\rho_{\mathbf{G}}, \tag{2}$$

where $\mathbf{G}$ and $\mathbf{G}'$ are the reciprocal lattice vectors. The Eq. 2 can be rewritten as

$$\sum_{\mathbf{G}'} \varepsilon^{\mathrm{M}}_{\mathbf{G}\mathbf{G}'}V_{\mathbf{G}'} = 4\pi\rho_{\mathbf{G}}/G^2, \tag{3}$$

$$\varepsilon^{\mathrm{M}}_{\mathbf{G}\mathbf{G}'} \equiv \varepsilon^{\mathrm{M}}_{\mathbf{G}-\mathbf{G}'}\mathbf{G}\cdot\mathbf{G}'/G^2. \tag{4}$$

Here, we define the macroscopic static dielectric matrix, $\varepsilon^{\mathrm{M}}_{\mathbf{G}\mathbf{G}'}$. On the other hand, the static dielectric matrix $\varepsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q};0)$ of a system in vacuum is related to its static polarizability $\chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q};0)$ as

$$\varepsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q};0) = \delta_{\mathbf{G}\mathbf{G}'} - v(\mathbf{q}+\mathbf{G})\chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q};0), \tag{5}$$

where $v(\mathbf{q}+\mathbf{G})$ is the bare Coulomb interaction. When the system is nearby a dielectric media, the additional screening by the dielectrics can be approximated by the macroscopic dielectric polarizability, $\chi^{\mathrm{M}}_{\mathbf{G}\mathbf{G}'}(\mathbf{q};0)$. $\chi^{\mathrm{M}}_{\mathbf{G}\mathbf{G}'}(\mathbf{q};0)$ is then added to the static polarizability $\chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q};0)$ of the main system of interest:

$$\varepsilon_{\mathbf{G}\mathbf{G}'}(\mathbf{q};0) = \delta_{\mathbf{G}\mathbf{G}'} - v(\mathbf{q}+\mathbf{G})\{\chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q};0) + \chi^{\mathrm{M}}_{\mathbf{G}\mathbf{G}'}(\mathbf{q};0)\} \tag{6}$$

$$= \varepsilon^{\mathrm{M}}_{\mathbf{G}\mathbf{G}'}(\mathbf{q};0) - v(\mathbf{q}+\mathbf{G})\chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q};0). \tag{7}$$

In Eq. 7, the only difference from the static dielectric matrix of Eq. 5 is $\varepsilon^{\mathrm{M}}_{\mathbf{GG'}}(\mathbf{q};0)$ instead of $\delta_{\mathbf{GG'}}$, which represents that the vacuum is replaced by the dielectric media where the system is located. The $\varepsilon^{\mathrm{M}}_{\mathbf{GG'}}(\mathbf{q};0)$ can be obtained from Eq. 4 for the $|\mathbf{q}\rangle$ electronic state as

$$\varepsilon^{\mathrm{M}}_{\mathbf{GG'}}(\mathbf{q};0) = \varepsilon^{\mathrm{M}}_{\mathbf{q+G,q+G'}} = \varepsilon^{\mathrm{M}}_{\mathbf{G-G'}}(\mathbf{q}+\mathbf{G})\cdot(\mathbf{q}+\mathbf{G'})/|\mathbf{q}+\mathbf{G}|^{\mathbf{2}}. \quad (8)$$

The real-space macroscopic dielectric constant profiles $[\varepsilon^{\mathrm{M}}(\mathbf{r})]$ along the perpendicular direction to the ML MoS_2 are shown in Fig. S1 for the model structures of (a) the ML MoS_2 with one-side HfO_2, (b) the ML MoS_2 with both-side HfO_2, (c) the ML MoS_2 with one-side Au, and (d) the ML MoS_2 with both-side Au. The dielectric interfaces are chosen to be the top- and bottom-most atomic plane of the HfO_2 and Au slabs. At the interfaces, the dielectric profiles were smoothened with the error functions (β=0.2 Å).

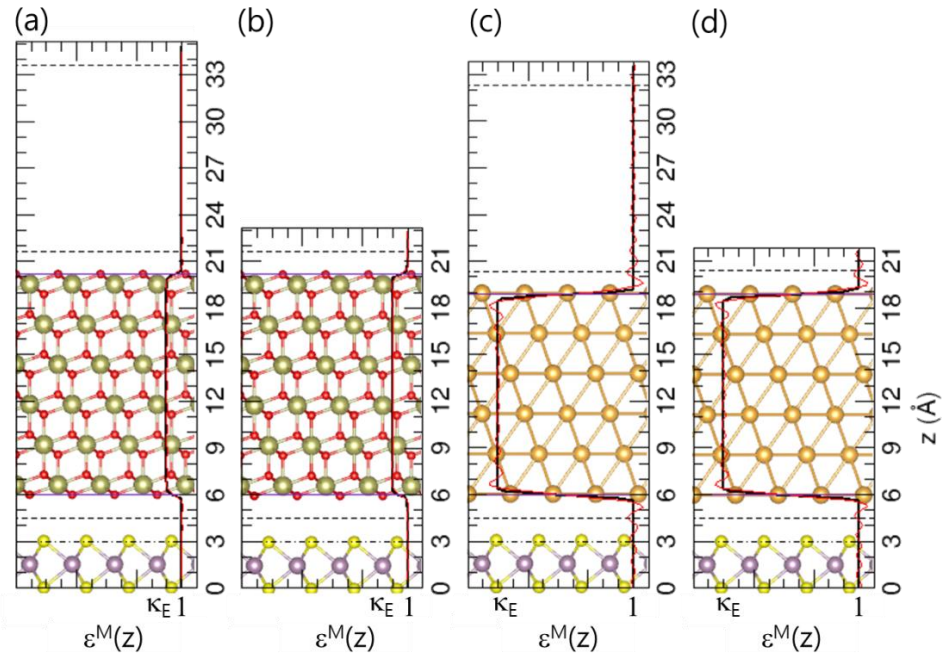

Figure S1. **Real-space environmental dielectric constant profiles.** (a-d) The dielectric constant profiles along the perpendicular direction to the ML MoS_2 for (a) the ML MoS_2 with

one-side HfO_2, (b) the ML MoS_2 with both-side HfO_2, (c) the ML MoS_2 with one-side Au, and (d) the ML MoS_2 with both-side Au.