Electronic Supplementary Information

Two-dimensional β-phase group-VA binary compounds for versatile electronic and optical properties

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S1. Formation energy

The formation energy of binary monolayers is calculated by $\Delta E_{\mathrm{f}} = E(\mathrm{AB}) - \frac{1}{2}\left[E(\mathrm{A}) + E(\mathrm{B})\right]$, where $E(\mathrm{AB})$ represents the total energy per each atom of binary monolayers, and $E(\mathrm{A})$ and $E(\mathrm{B})$ represent those of elemental monolayers.

S2. Formation energy

The cohesive energy of binary monolayers is calculated by $\Delta E_{\mathrm{c}} = E(\mathrm{AB}) - \frac{1}{2}\left[E'(\mathrm{A}) + E'(\mathrm{B})\right]$, where $E(\mathrm{AB})$ represents the total energy per each atom of binary monolayers, and $E'(\mathrm{A})$ and $E'(\mathrm{B})$ represent those of isolated elemental A and B atoms.

S3. Deformation charge density

Deformation charge density of system AB is defined as $\Delta\rho = \rho_{\mathrm{AB}} - \rho_{\mathrm{A}} - \rho_{\mathrm{B}}$, where ρ_{AB} is the charge density of monolayer AB and ρ_{A} or ρ_{B} is the atomic charge density of the A or B atoms in the monolayer AB.

S4. carrier effective mass and mobility

The carrier effective masses are calculated by fitting the conventional equation $E(\mathbf{k}) = E_0 + \hbar^2\mathbf{k}^2/2m^*$, where E_0 is the band energy at the k-point and $\mathbf{k}$ is the wave-vector relative to the k-point[1].

The carrier mobilities (μ) were calculated using the widely used deformation potential (DP) theory,[2] with $\mu = \frac{e\hbar^3 C_{2D}}{K_B T m_i^* m_d (E_1)^2}$. Here, e is the elementary charge, $\hbar$ the reduced Planck constant, K_B the Boltzmann constant, T the temperature, m_i^* the carrier effective mass along the transport direction, m_d the average effective mass defined as $m_d = (m_x^* m_y^*)^{\frac{1}{2}}$, C_{2D} the elastic modulus, and E_1 deformation potential. The x and y are the special zigzag and armchair transport directions, as shown in Fig. 1. To obtain the C_{2D} and E_1, four strains of δ (-1.00%, -0.05%, 0.05% and 1.00%) are applied on the perfect 2D lattice. The elastic modulus C_{2D} is fitted as per the response of total energy toward the applied strains by $C_{2D} = 2\Delta E/(\delta^2 S_0)$, where ΔE is the total energy change due to the applied strains and S_0 is the area of the 2D supercell at the equilibrium state. The deformation potential is fitted by $E_1 = \Delta E_{\mathrm{b}}/\delta$, where ΔE_{b} is the energy changes of the band edges (the top of valence band for holes and the bottom of the conduction band for electrons) due to the applied strains.

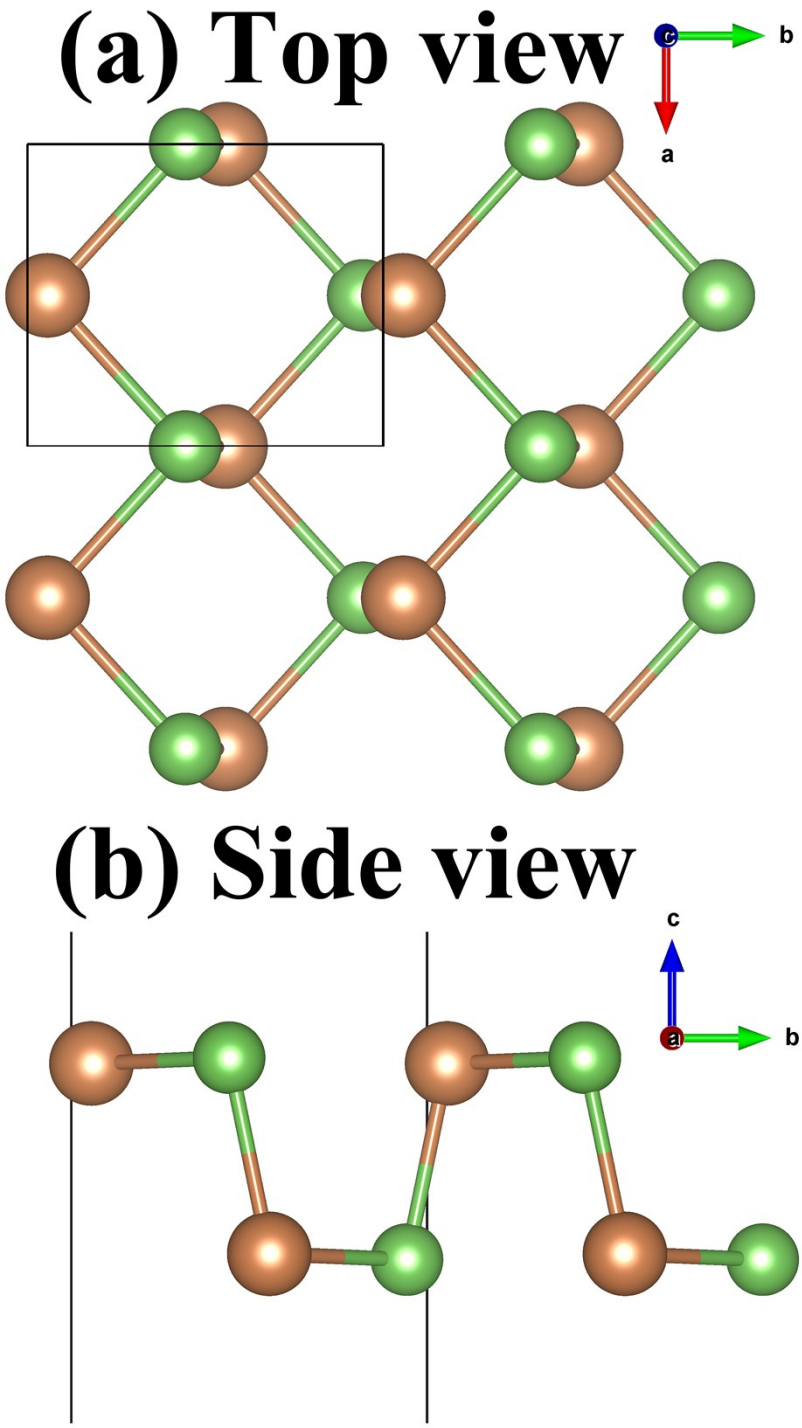

Fig. S1 (a) Top and (b) side views of atomistic structure of 2D α-AsSb monolayer. The As and Sb atoms are represented by green and brown balls.

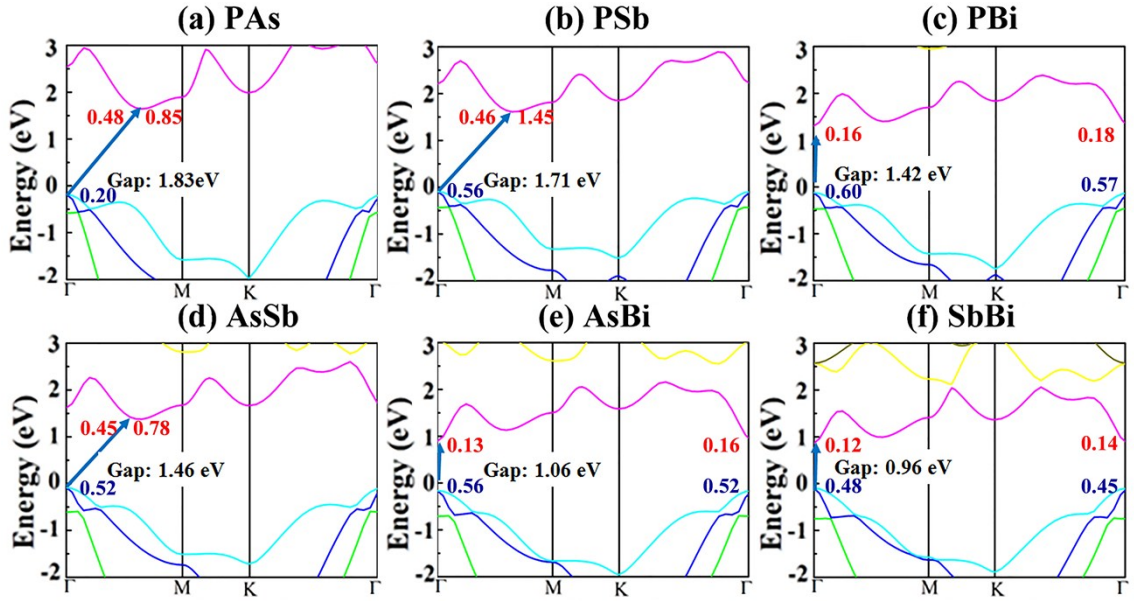

Fig. S2 Band structures for 2D β-phase binary group-VA monolayers at the PBE level. The electron and hole effective masses for different directions are labeled in red and blue, respectively. The effective masses are given in the unit of the free-electron mass m_0.

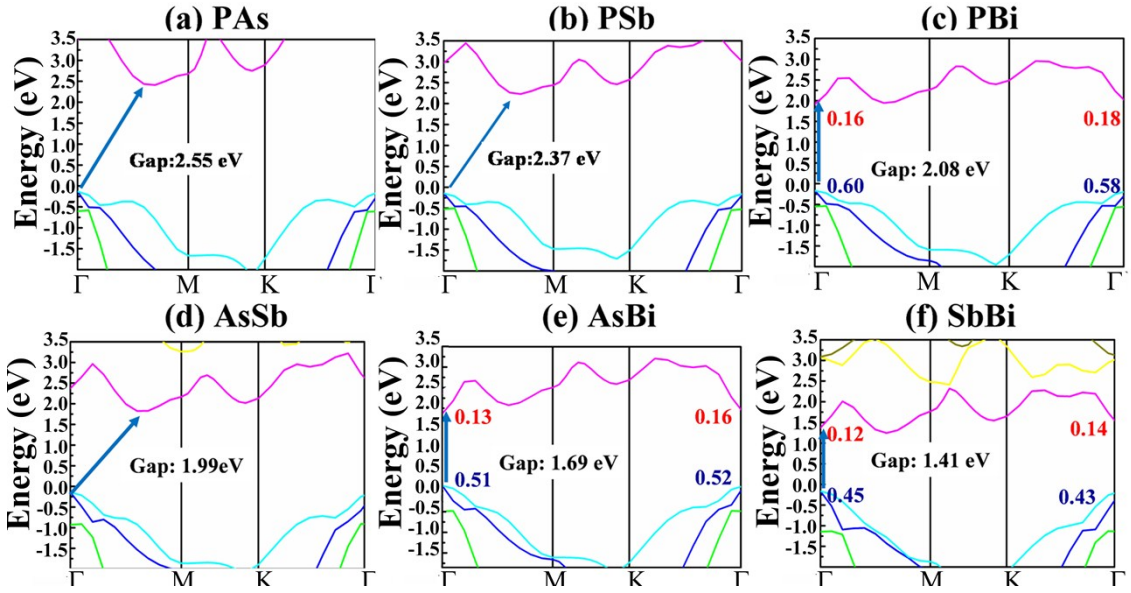

Fig. S3 Band structures for 2D β-phase binary group-VA monolayers at the HSE06 level. The electron and hole effective masses of bismuthides for different directions are labeled in red and blue, respectively. The effective masses are given in the unit of the free-electron mass m_0.

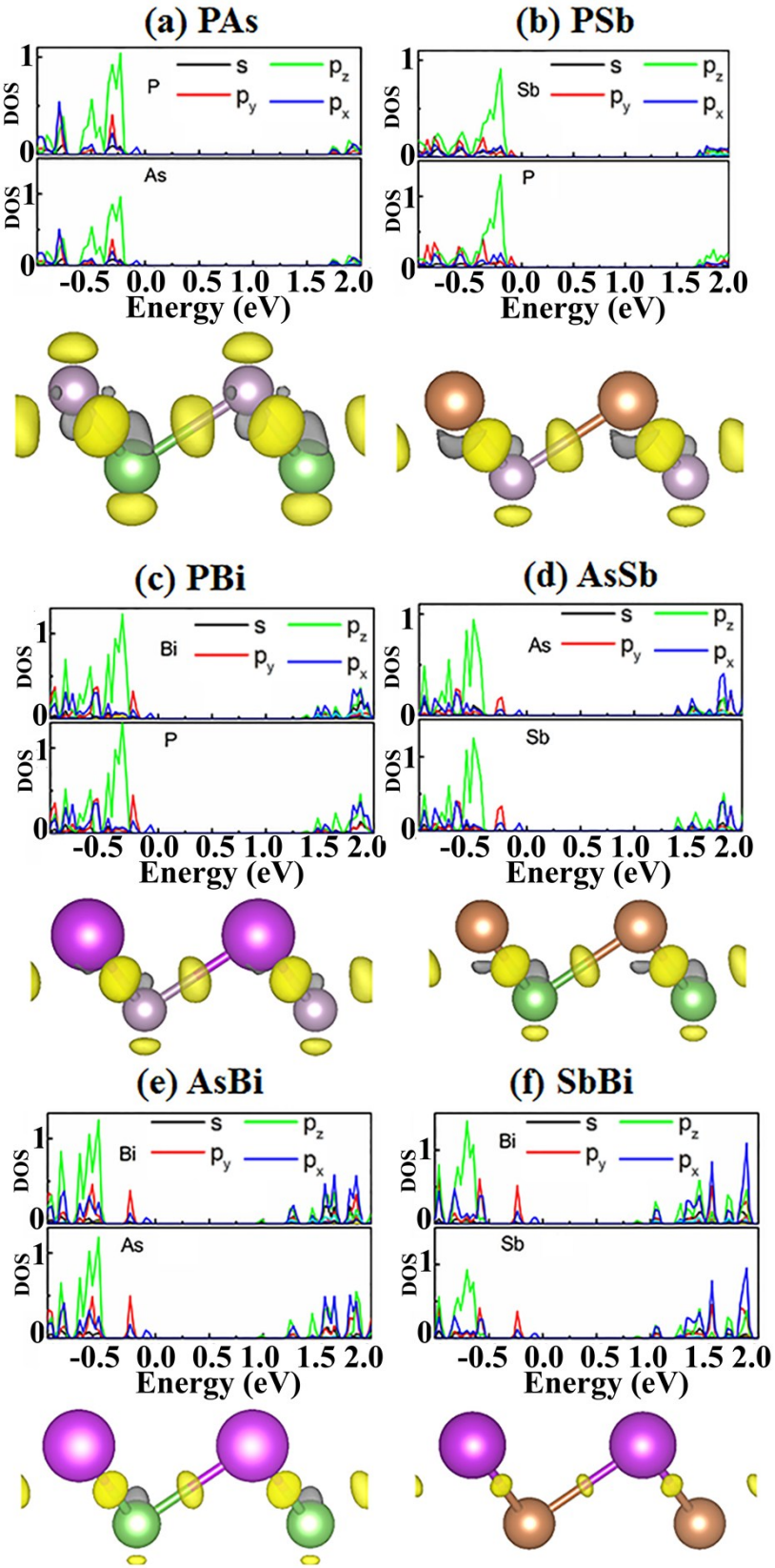

Fig. S4 Projected density of states (DOS) and deformation charge density of 2D β-phase binary group-VA monolayers at the PBE level. Charge accumulation and depletion with respect to isolated atoms are also depicted in yellow and gray. The isosurface is at 0.01 eÅ^{-3}.

Table S1 effective mass m^*/m_0, DP constant E_1 in eV, elastic constant C_{2D} in Jm^{-2}, and carrier

mobility μ in $cm^{-2}V^{-1}s^{-1}$, at 300 K for electrons (e) and holes (h) in 2D β-phase monolayer bismuthides. The results are calculated with the PBE functional.

Materials		m_x^* / m_0 [a]	m_y^* / m_0 [a]	E_{lx}	E_{ly}	$C_{x/2D}$	$C_{y/2D}$	μ_x	μ_y
PBi	e	0.18	0.16	9.79	9.77	44.63	39.46	306.00	324.43
	h	0.57	0.60	6.96	6.89	44.63	39.46	58.79	49.11
AsBi	e	0.16	0.13	8.89	8.85	39.23	34.83	413.26	504.74
	h	0.52	0.56	7.15	7.05	39.23	34.83	58.26	47.55
SbBi	e	0.12	0.14	8.83	8.80	31.56	27.90	598.91	422.54
	h	0.48	0.45	7.67	7.57	31.56	27.90	51.16	51.15

[a] m_0 is the mass of free electron

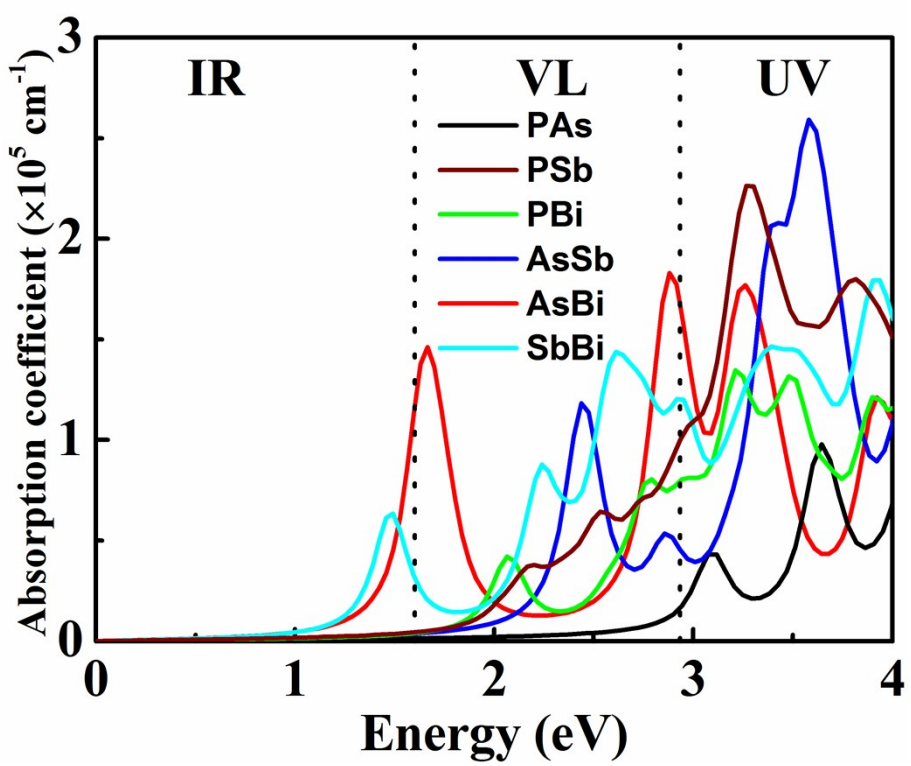

Fig. S5 Calculated optical absorption spectra at the HSE06 level for 2D β-phase binary group-VA monolayers. The energy range for the IR (Infra-red), VL (visible light) and UV (ultraviolet) regions are represented by vertical lines.

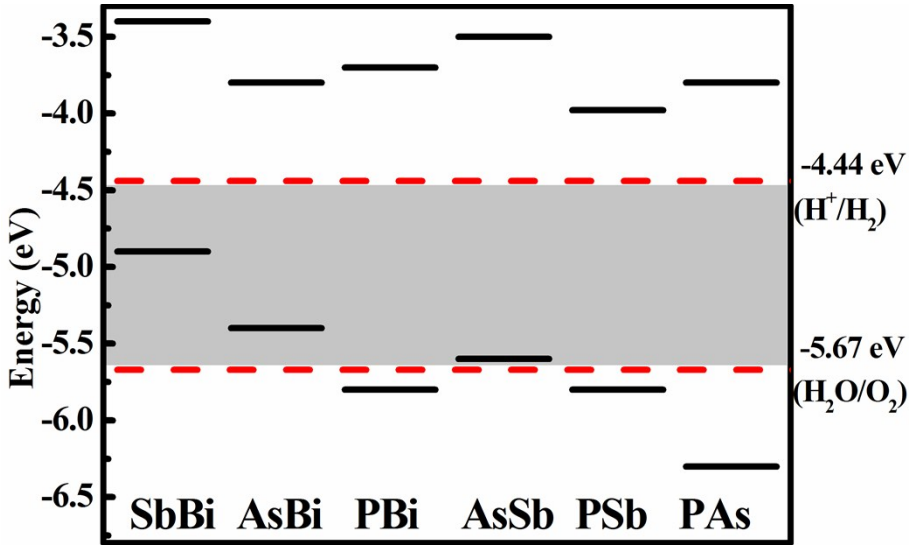

Fig. S6 Band alignments of CBM (upper) and VBM (lower) for 2D β-phase binary group-VA monolayers with HSE06. The dotted lines indicate the water reduction (H^+/H_2) and oxidation (H_2O/O_2) potentials at pH=0.

References:

1 J. Y. Xi, T. Q. Zhao, D. Wang and Z. G. Shuai, *J. Phys. Chem. Lett.*, 2013, **5**, 285.
2 M. Q. Xie, S. L. Zhang, B. Cai, Z. Zhu, Y. S. Zou and H. B. Zeng, *Nanoscale*, 2016, **8**, 13407.