Supporting Information of

The Optimal Electronic Structure for High-Mobility 2D Semiconductors: Exceptionally High Hole Mobility in 2D Antimony

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Computational Methods

We perform first-principles calculations using the Quantum Espresso Package[1] with the SG15 Optimized Norm-Conserving Vanderbilt pseudopotentials (SG15 ONCV Potentials) [2-4] and the Perdew-Burke-Ernzerhof[5] (PBE) exchange-correlation functional. The kinetic energy cutoffs for wave functions and charge density are set to 60 and 240 Ry, respectively. The atomic coordinates are optimized using a 36 × 36 $\boldsymbol{k}$ mesh until the magnitude of the force acting on each atom becomes less than 0.0001 Ry/Bohr. The spin-orbit coupling is included for As, Sb, and Bi.

To obtain the mobility, we first use density functional perturbation theory[6] (DFPT) to calculate the electron-phonon coupling (EPC) strengths (g) for 9 × 9 $\boldsymbol{k}$ (electron wavevector) points and 9 × 9 $\boldsymbol{q}$ (phonon wavevector) points, where three (six for the spin-orbit case) p orbitals per atom is used for the initial Wannier projection[7]. Then the g for 300 × 300 $\boldsymbol{k}$ and 300 × 300 $\boldsymbol{q}$ points are obtained using the interpolation approach as implemented in the EPW code[8]. We use the Gaussian function with a broadening of 5 meV to account for the Delta function in Eq. 2. The lower boundary for the phonon frequency in the electron-phonon coupling calculations is set as 5 cm^{-1} (corresponds to 0.62 meV). We find that 300 × 300 $\boldsymbol{k}$ and $\boldsymbol{q}$ are sufficient to converge the mobility values.

The contribution of an electronic state $i\mathbf{k}$ to the mobility is characterized as:

$$p(i\mathbf{k}) = \frac{\tau(i\mathbf{k})v^2(i\mathbf{k})\left|\frac{\partial f_0}{\partial E_{i\mathbf{k}}}\right|}{\sum_i \int_{BZ} f_0 d\mathbf{k}}, \qquad \text{(S1)}$$

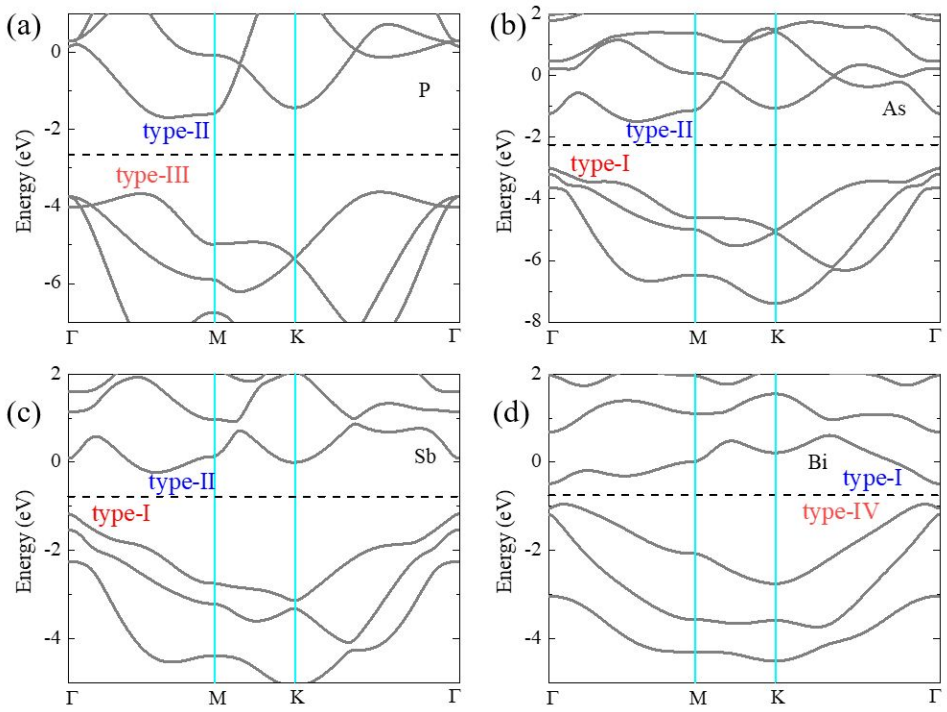

Figure S1 Electronic band structures of the group-15 hexagonal 2D materials. The Fermi levels are shown by the black dashed lines. The band types near the band edge states are labelled.

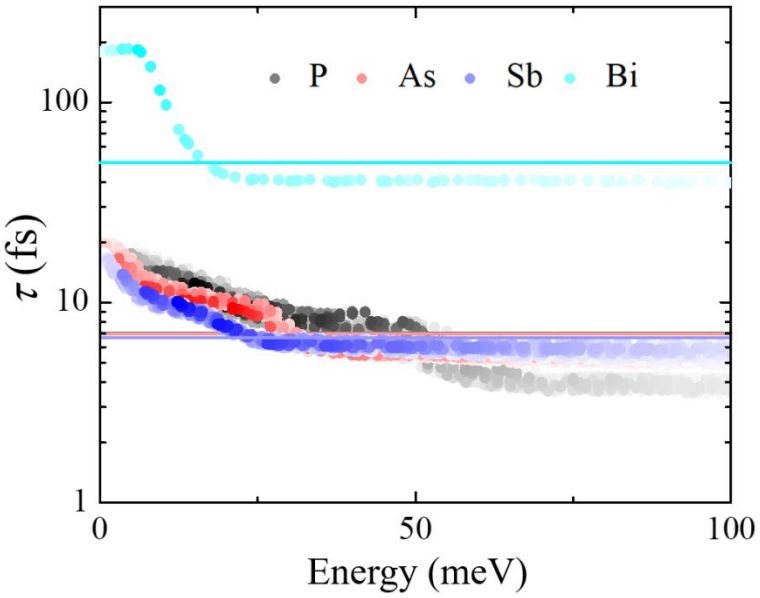

Figure S2 Relaxation time τ of the electrons (shown by dots), and the "average relaxation time" $\bar{\tau}$ (shown by lines). The energy of the conduction band minimum is set to be zero. The color shade of the τ represents the contribution of the corresponding electronic state to the mobility.

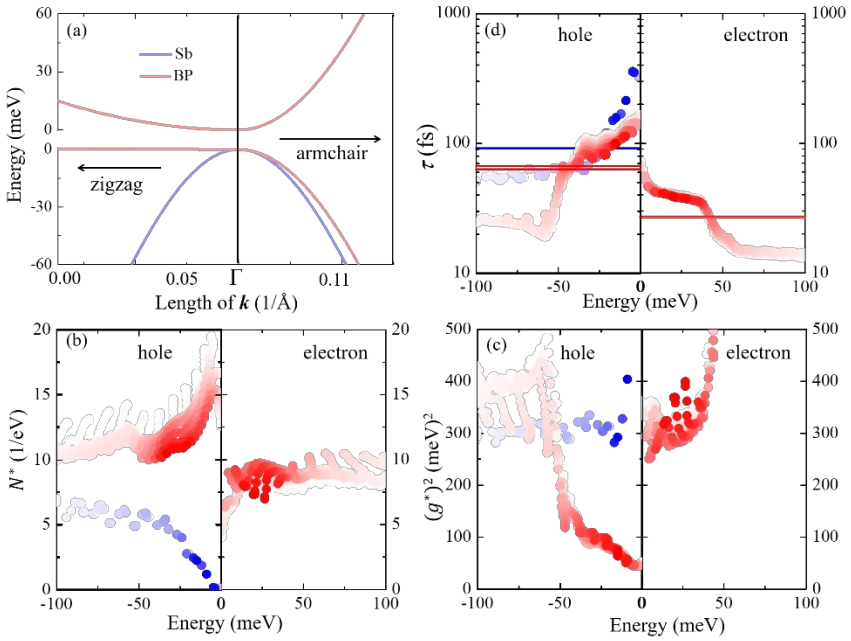

Figure S3 (a) Band structure of monolayer black phosphorus (BP) and Sb. (b-d) Comparison of effective number of states (b), effective EPC strength (c), and relaxation time (d), between the BP (electrons and holes) and Sb (holes). The average relaxation times are shown in (d) as horizontal lines. The color strength represents the contribution of the corresponding electronic state to the mobility.

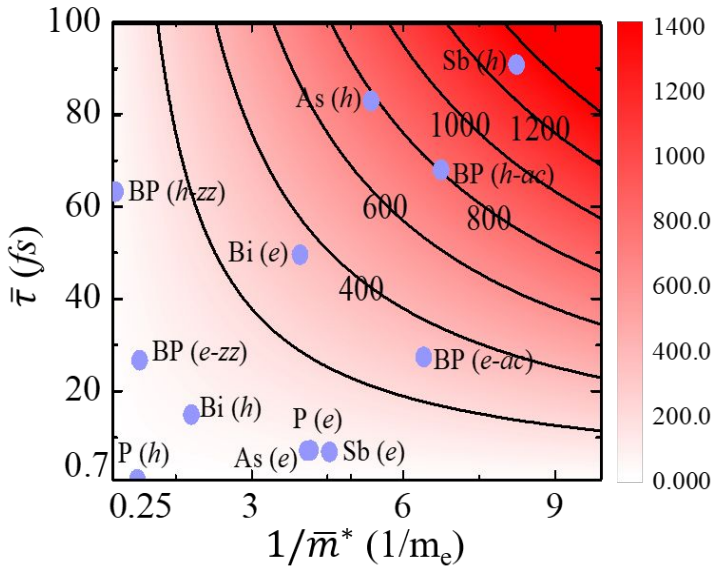

Figure S4 Carrier mobility as a function of "average effective mass" $\bar{m}^*$ and "average relaxation time" $\bar{\tau}$ (see the main text for definition). Note that the BP has an anisotropic structure (rectangular) while others have isotropic (hexagonal) structures.

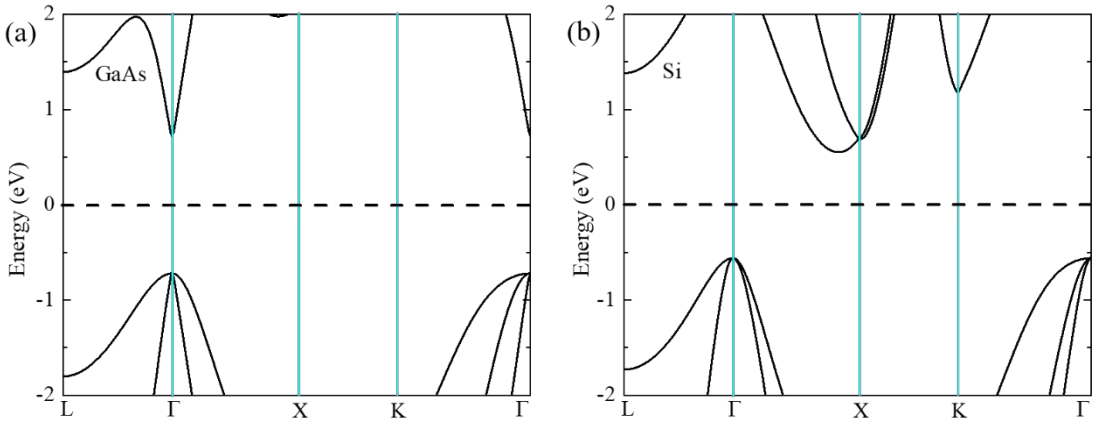

Figure S5 Electronic structures of GaAs (a), and Si (b), calculated using PBE functional with the band gap shifted to be experimental value.

Table S1 The effective mass m^* of the band edge state, the "average effective mass" $\bar{m}^*$, and the "average relaxation time" $\bar{\tau}$ of the group-15 2D hexagonal materials.

	holes			electrons		
	m^* (m_e)	$\bar{m}^*$ (m_e)	$\bar{\tau}$ (fs)	m^* (m_e)	$\bar{m}^*$ (m_e)	$\bar{\tau}$ (fs)
2D P	1.3613	1.3400	0.7	0.2432	0.2383	7.1
2D As	0.1549	0.1850	83.2	0.2353	0.2420	7.0
2D Sb	0.1050	0.1204	91.2	0.2162	0.2187	6.7
2D Bi	0.4443	0.5489	14.8	0.2343	0.2507	49.7

Table S2 Comparison between experimentally measured and theoretically calculated (using Boltzmann transport theory with scattering rates obtained from first principles) electron mobility at room temperature for Si, GaAs, and monolayer MoS_2. Note that for monolayer MoS_2, we choose the experiments that use four-terminal or Hall measurements so that the contact resistance is subtracted.

	Experimental ($cm^2V^{-1}s^{-1}$)	Calculation ($cm^2V^{-1}s^{-1}$)
Si (e)	1300 ~ 1450, Ref.[9-11]	1366, Ref.[12]
GaAs (e)	8200 ~ 8900, Ref.[13-19]	8899, Ref.[20]
Monolayer MoS_2 (e)	11 ~ 165, Ref.[21-23]	176, this work

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