Electronic Supplementary Information

Semiconducting two-dimensional group VA-VA haeckelite compounds with superior carrier mobility

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Figure list:

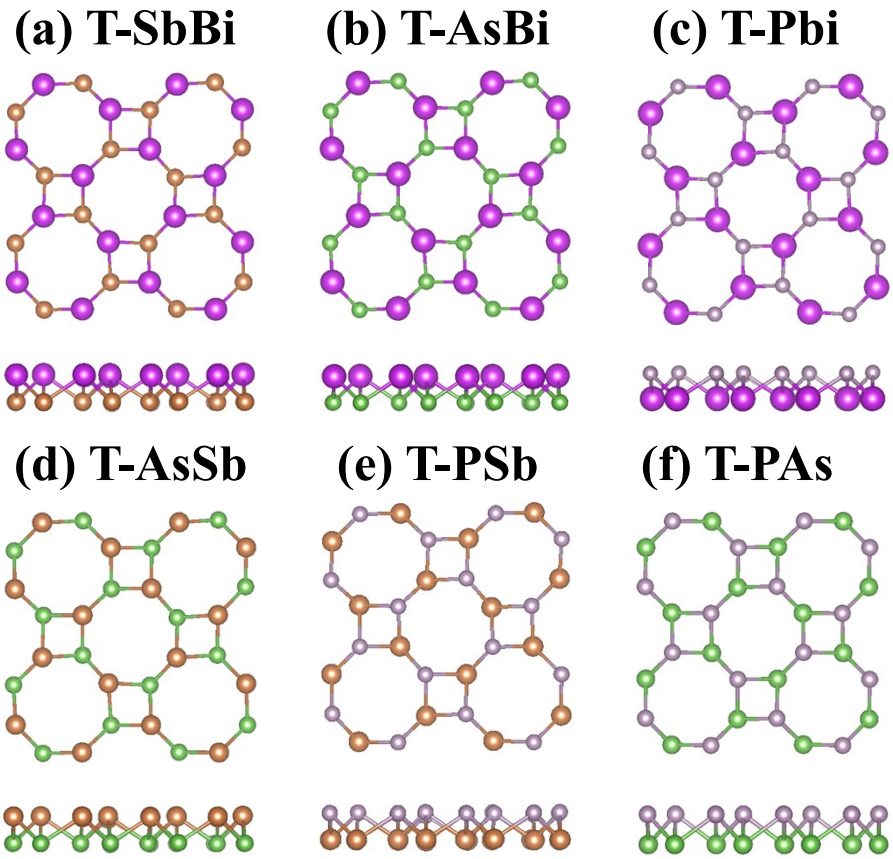

Fig. S1 Snapshots of the final frame of 2D T-VA-VAs at 300K at the end of 5 ps ab-initio molecular dynamics (AIMD) simulations.

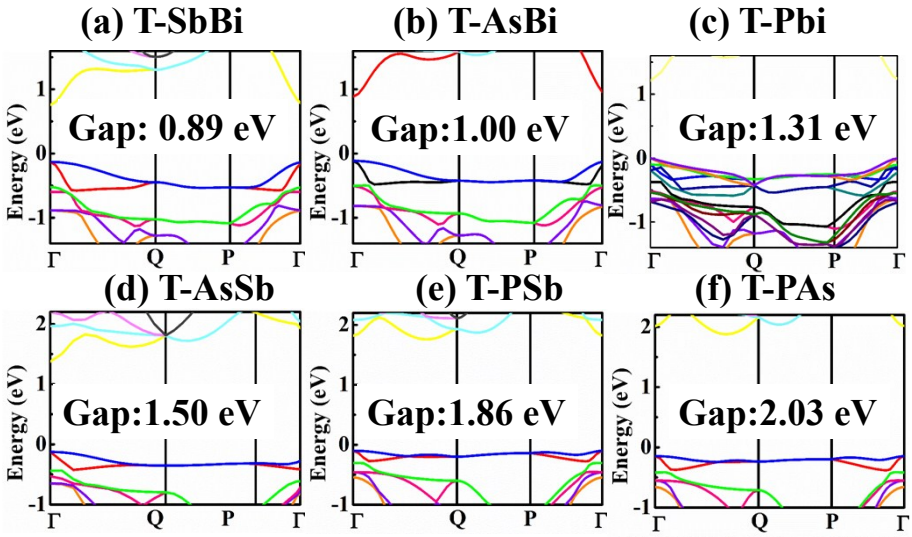

Fig. S2 Band structures for 2D binary T-VVs with PBE level.

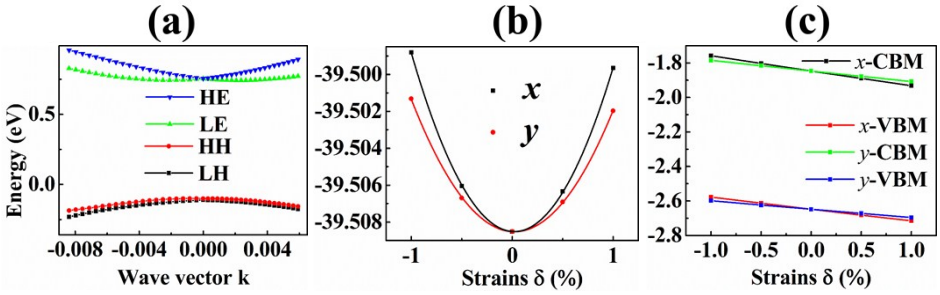

Fig. S3 Effective mass, 2D elastic constant and DP constant evaluation for 2D binary T-SbBi, which are calculated with HSE+SOC. (a) the band energies with respect to *k* vectors. The quadratic fit of the data gives the effective masses. (b) Total energies with respect to the lattice strains of δ. The quadratic fit of the data gives the 2D elastic constant. (c) Band energies of the VBMs and CBMs with respect to the vacuum energy as a function of. The linear fit of the data gives the DP constant.

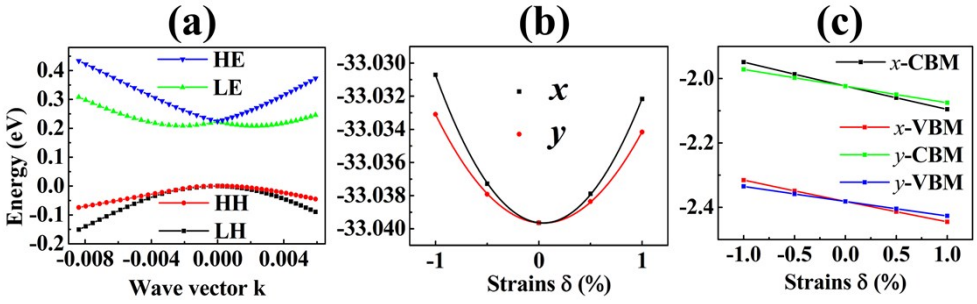

Fig. S4 The same results as in Fig. S3 but are calculated from PBE+SOC.

Table list:

Table S1 Effective mass in the mass of free electron m_0, DP constant E_1 in eV, elastic constant C_{2D} in Jm^{-2}, and carrier mobility μ in $10^3 cm^{-2}V^{-1}s^{-1}$, at 300 K for electrons (e) and holes (h) for direct semiconductors with haeckelite structure. The results are calculated with PBE.

Materials		m_x^*	m_y^*	E_{1x}	E_{1y}	$C_{x/2D}$	$C_{y/2D}$	μ_x	μ_y
SbBi	h	1.53	0.33	6.67	4.69	40.29	29.79	0.18	0.27
	e	0.07	0.07	8.22	5.81	40.29	29.79	2.87	4.38
AsBi	h	1.63	0.38	6.67	3.73	47.65	20.08	0.02	0.21
	e	0.07	0.07	8.54	4.49	47.65	20.08	2.51	3.93
PBi	h	1.87	0.46	6.14	4.31	96.96	70.12	0.02	0.21
	e	0.09	0.09	9.63	6.81	96.96	70.12	1.39	2.05
AsSb	h	1.80	0.42	6.68	4.69	52.02	25.60	0.02	0.14
	e	0.10	0.09	9.88	6.99	52.02	25.60	1.28	1.30

Table S2 Details on effective mass with HSE+SOC. The effective masses of holes are calculated by the average value of the LH and HH as $m^h = (m^{\mathrm{LH}} m^{\mathrm{HH}})^{\frac{1}{2}}$, while those of electrons are calculated as $m^e = (m^{\mathrm{LE}} m^{\mathrm{HE}})^{\frac{1}{2}}$. The unit of effective mass is in the mass of free electron m_0

Materials		m^{LH}	m^{HH}	m^h	m^{LE}	m^{HE}	m^e
SbBi	x	0.14	0.09	0.11	0.07	0.10	0.08
	y	0.11	0.10	0.10	0.07	0.08	0.08
AsBi	x	0.17	0.13	0.14	0.22	0.06	0.13
	y	0.02	0.12	0.15	0.21	0.06	0.11
PBi	x	0.24	0.18	0.21	0.48	0.18	0.29
	y	0.30	0.17	0.23	0.33	0.23	0.28
AsSb	x	0.14	0.13	0.13	0.12	0.89	0.10
	y	0.15	0.15	0.15	0.11	0.10	0.10

Table S3 The same results as in Table S1 but are calculated with HSE+SOC.

Materials		m_x^*	m_y^*	E_{1x}	E_{1y}	$C_{x/2D}$	$C_{y/2D}$	μ_x	μ_y
SbBi	h	0.11	0.10	6.89	4.87	41.74	30.89	1.67	2.71
	e	0.08	0.08	8.70	6.15	41.74	30.89	1.89	2.96
AsBi	h	0.14	0.15	6.16	4.38	51.21	37.26	1.36	1.89
	e	0.13	0.11	9.13	6.45	51.21	37.26	1.03	1.53
PBi	h	0.21	0.23	6.04	4.27	57.26	41.50	0.76	1.00
	e	0.29	0.28	9.93	7.02	51.21	37.26	0.15	0.23
AsSb	h	0.13	0.15	7.08	5.00	60.28	44.56	1.33	1.76
	e	0.10	0.10	10.77	7.62	51.21	37.26	1.06	1.55

S1. POSCAR of T-SbBi optimized by VASP

T-SbBi

1.00000000000000

8.3080340759988136 0.0000000000000000 0.0000000000000000
0.0000000000000000 8.3080309430284558 0.0000000000000000
0.0000000000000000 0.0000000000000000 20.0000000000000000
Sb Bi
4 4
Direct
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0.1440782650773168 0.6440783732483405 0.4584629994970427
0.8559221344902278 0.3559212862063713 0.4584633196217811
0.3499557996682872 0.8499555386322363 0.5415368584308758
0.6500443121165681 0.1500440171565955 0.5415368169112336
0.8499564113675797 0.6500443878051320 0.5415366132051276
0.1500436971232153 0.3499563057850587 0.5415369135823312

S2. Cohesive energy

The cohesive energy of binary monolayers is calculated by $\Delta E_c = E(\mathrm{AB}) - \frac{1}{2}\left[E'(\mathrm{A}) + E'(\mathrm{B})\right]$, 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. Carrier effective mass and mobility

The band energy nearby the VBM or CBM with wave-vector *k* are fitted by the conventional equation $E(k) = E_0 + \hbar^2 k^2 / 2m^*$, where E_0 is the band energy value of VBM or CBM. Then effective mass[1] of the hole or electron is calculated as $m^* = \hbar^2 / \frac{\partial^2 E(k)}{\partial k^2}$. Both the CBM and VBM are nondegenerate due to SOC effect, as shown in Fig. 3. Thus, the hole effective masses is calculated by the average value of low hole (LH) and high hole (HH), and the electron effective masses is calculated by the average value of low electron (LE) and high electron (HE).

S4. Carrier mobility

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 $E_1 = \Delta E_b / \delta$, 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.

S5. Optical absorption

The optical absorption coefficients[3] are calculated from the complex dielectric function $I(\omega) = \sqrt{2}\omega(\sqrt{\varepsilon'(\omega)^2 + \varepsilon''(\omega)^2} - \varepsilon'(\omega))^{1/2}$, where $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ are the real and imaginary parts of the complex dielectric constant at absorption frequency ω.

References:

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3. S. Sonali, T. P. Sinha and M. Abhijit, *J. Phys.: Condens. Matter*, 2000, **12**, 3325.