Two-dimensional AlXY (X= S, Se and Y = Cl, Br, I) monolayers: promising photocatalysts for water splitting with high anisotropic carrier mobilities

Lu Pan [1], Tian Zhang [2], Cui-E Hu [3,*], Xiang-Rong Chen [1,*], Hua-Yun Geng [4]

[1] College of Physics, Institute of Atomic and Molecular Physics, Sichuan University, Chengdu 610065, China;

[2] College of Physics and Electronic Engineering, Sichuan Normal University, Chengdu 610066, China;

[3] College of Physics and Electronic Engineering, Chongqing Normal University, Chongqing 400047, China;

[4] National Key Laboratory for Shock Wave and Detonation Physics Research, Institute of Fluid Physics, CAEP, Mianyang 621900, China

ABSTRACT

We report a new group of two-dimensional (2D) AlXY (X= S, Se and Y = Cl, Br, I) monolayers by means of first-principles calculations. These AlXY monolayers were found to possess favorable dynamic, thermal and mechanical stabilities. The calculated Young's modulus and Poisson's ratio reflect significant anisotropy in the mechanical properties of AlXY monolayers. Our electronic calculations indicate that these monolayers exhibit semiconductor character and their bandgaps range from 2.25 eV to 3.31 eV. Among these monolayers, we find that four monolayers, namely AlSI, AlSeCl, AlSeBr and AlSeI, can be used as efficient photocatalysts. The AlSI monolayer exhibits a favorable photocatalytic activity in an acidic environment and the AlSeBr and AlSeI monolayers can serve as promising photocatalytic materials under acidic and neutral conditions. Notably, the AlSeCl monolayer shows the

* *Corresponding authors. E-mail*: cuiehu@cqnu.edu.cn; xrchen@scu.edu.cn

spontaneous overall photocatalytic water splitting in a wide range of pH conditions (0-14). Furthermore, the bandgaps and band edge positions of AlSCl, AlSBr, AlSI and AlSeBr monolayers can be tuned to fulfill highly efficient photocatalytic water splitting under an alkaline environment (pH = 14) by the appropriate external biaxial strains. The optical absorption and charge transport properties of AlXY monolayers are also investigated and the results reveal these monolayers exhibit strong absorption of ultraviolet light and high anisotropy in charge carrier transport. Therefore, the AlXY monolayers are expected to act as promising materials for photocatalytic water splitting.

Keywords: First-principles calculations; Anisotropic monolayers; Photocatalytic water splitting; Strain engineering;

Table S1 Calculated effective mass (m^*), average effective mass (m_d), elastic modulus (C_{2D}), deformation potential constant (E_1), carrier mobility (μ) and relaxation time (τ) for the AlXY (X= S, Se and Y = Cl, Br, I) monolayers at 300 K along the *x* and *y* directions.

	directions	carriers	$m^*(m_0)$	$m_d(m_0)$	C_{2D}	E_1(ev)	μ($cm^2V^{-1}s^{-1}$)	τ(10^{-14}s)
AlSCl	*x*	*e*	1.11	0.48	69.53	-1.97	716.62	45.29
		h	0.84	0.47	69.53	-7.97	60.37	2.89
	y	*e*	0.21	0.48	110.35	-15.24	100.45	1.20
		h	0.26	0.47	110.35	-12.95	114.75	1.70
AlSBr	*x*	*e*	0.89	0.41	63.79	-1.92	1010.61	51.21
		h	0.77	0.46	63.79	-8.53	52.75	2.31
	y	*e*	0.19	0.41	104.01	-16.03	110.73	1.20
		h	0.28	0.46	104.01	-11.20	137.19	2.19
AlSI	*x*	*e*	0.67	0.40	60.27	-3.35	427.06	16.29
		h	1.78	0.68	60.27	-3.57	83.26	8.44
	y	*e*	0.24	0.40	99.69	-14.44	106.13	1.45
		h	0.26	0.68	99.69	-13.69	64.11	0.95
AlSeCl	*x*	*e*	1.43	0.80	59.31	-4.23	61.75	5.03
		h	0.81	0.38	59.31	-7.63	70.54	3.25
	y	*e*	0.45	0.80	99.07	-4.77	257.76	6.60
		h	0.18	0.38	99.07	-14.50	176.17	1.81
AlSeBr	*x*	*e*	1.22	0.76	53.93	-4.43	63.16	4.39
		h	0.76	0.39	53.93	-7.98	60.89	2.63

AlSeI	*y*	*e*	0.48	0.76	93.56	-4.16	315.83	8.63
		h	0.20	0.39	93.56	-12.86	154.57	1.76
	x	*e*	0.82	0.37	49.10	-3.29	318.63	14.87
		h	0.66	0.38	49.10	-9.45	46.72	1.75
	y	*e*	0.17	0.37	88.75	-13.59	162.81	1.58
		h	0.22	0.38	88.75	-10.95	188.69	2.36

The parameters C_{2D}, m^* and E_l are defined as the following equations. $C_{2D} = (\partial^2 E/\partial \varepsilon^2)/S_0$, where E is the total energy under the uniaxial strain ε and S_0 is the area of the primitive cell. $m^* = \hbar^2[\partial^2 E_k/\partial k^2]^{-1}$, where E_k is the wave vector k dependent energy. $E_l = \partial E_{edge}/\partial \varepsilon$, where E_{edge} is value of CBM (electrons) or VBM (holes).

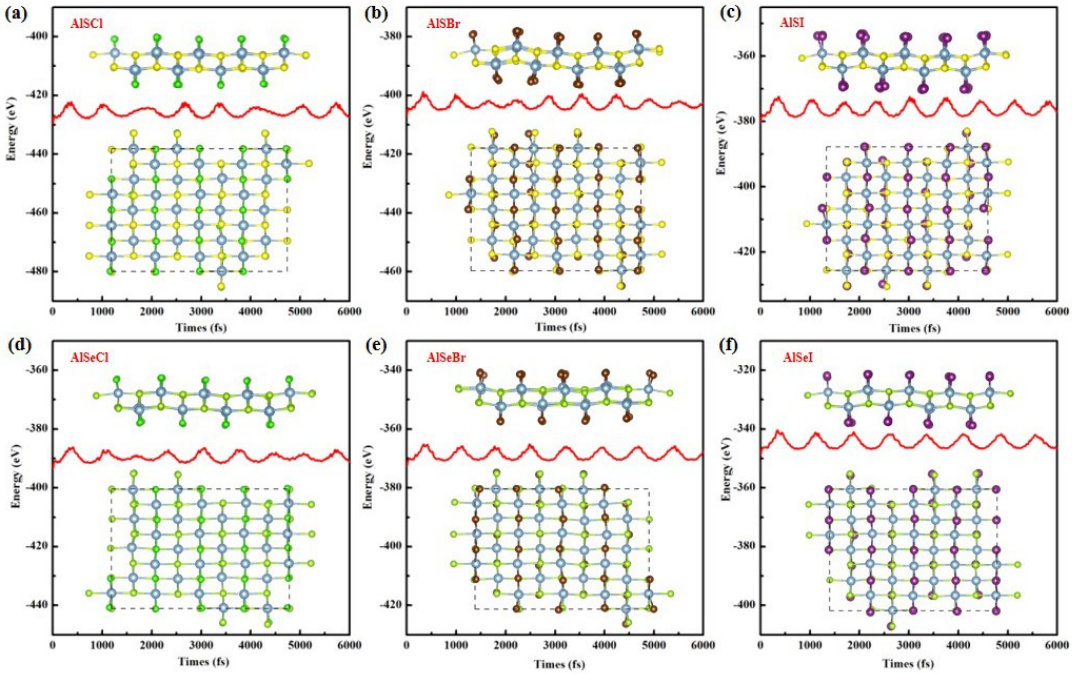

Fig. S1 (Color online) The structure snapshots from the AIMD simulations of the AlXY (X= S, Se and Y = Cl, Br, I) monolayers at 300 K, as well as the variation of the total energy.

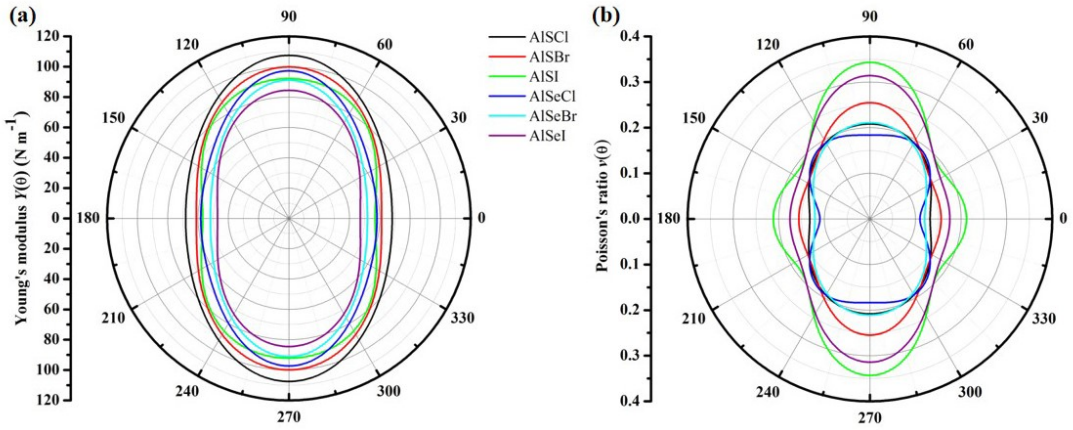

Fig. S2 (Color online) The orientation-dependent (a) Young's modulus $Y(\theta)$ and (b) Poisson's ratio $\nu(\theta)$ of the AlXY monolayers.

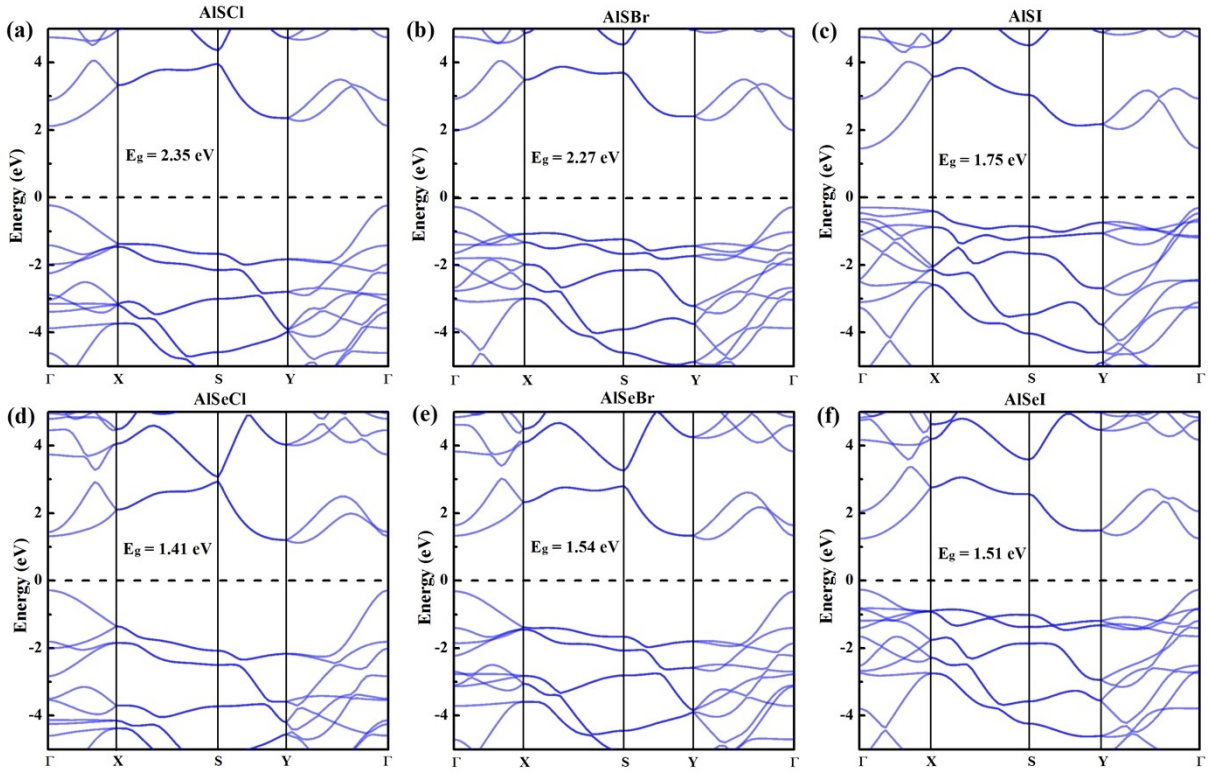

Fig. S3 (Color online) The calculated band structure of AlXY monolayers at the PBE level.

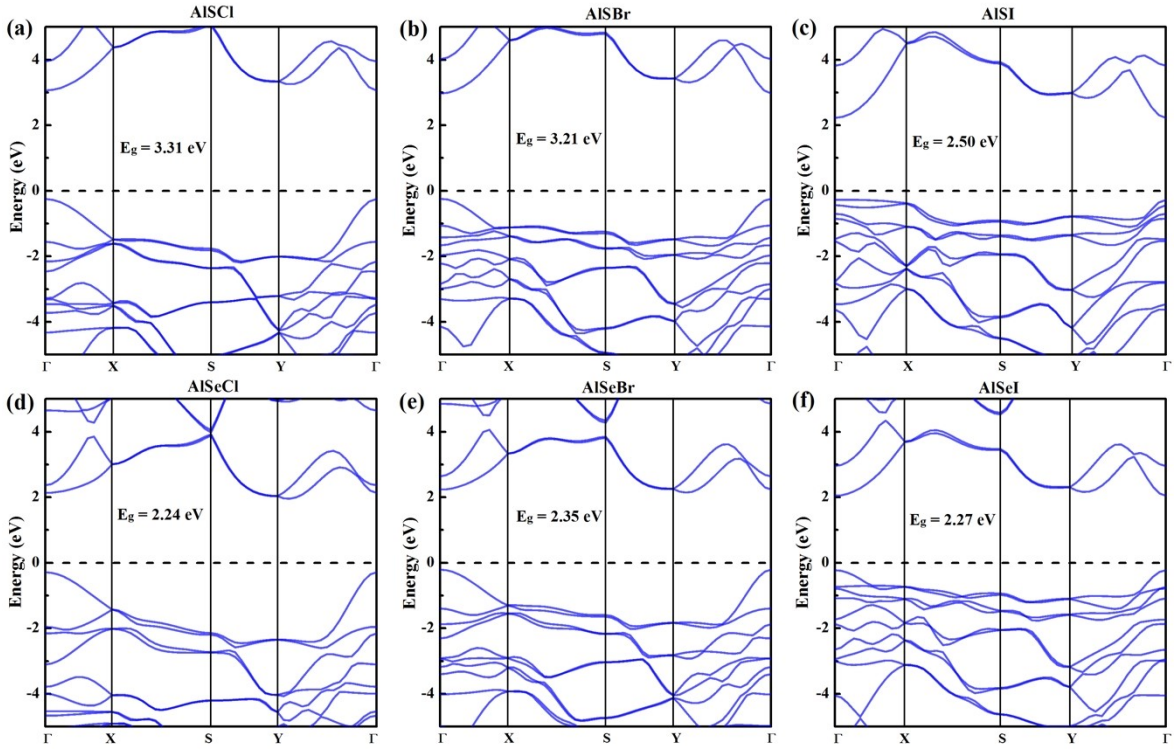

Fig. S4 (Color online) The calculated HSE06 band structure of AlXY (X= S, Se and Y = Cl, Br, I) monolayers with considering the SOC effect.

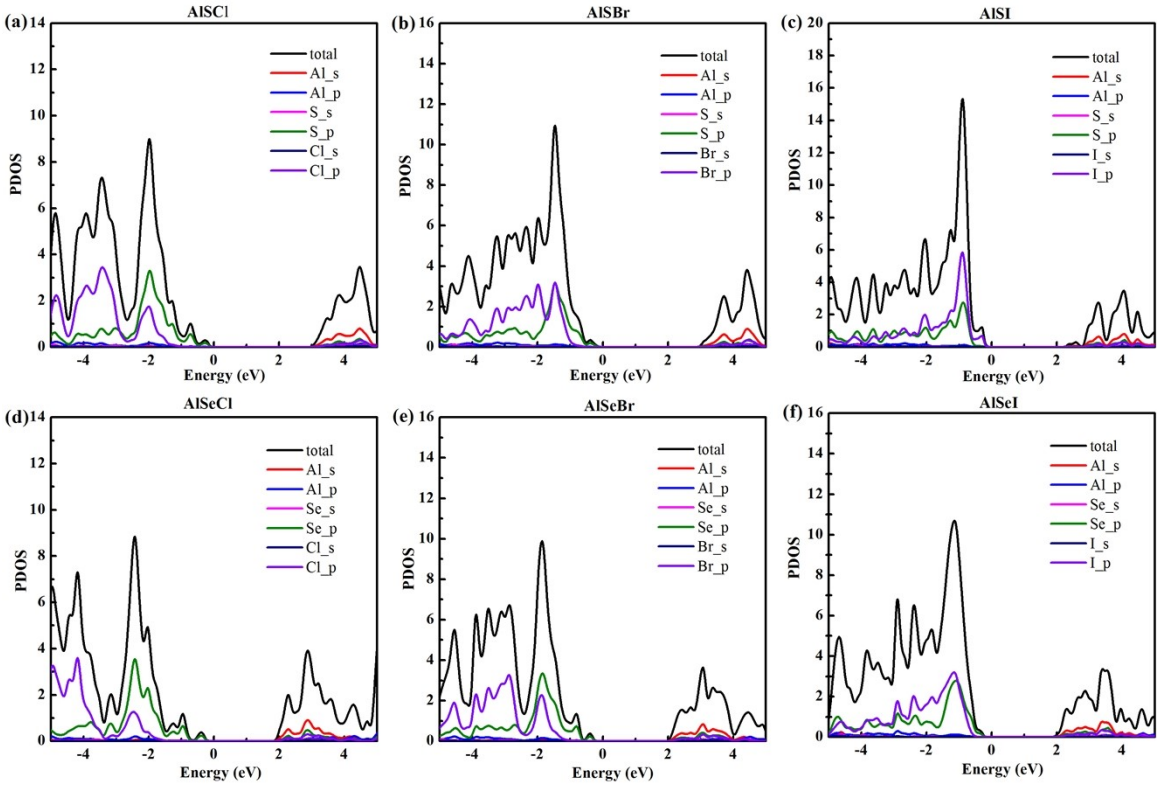

Fig. S5 (Color online) The calculated partial density of states (DOS) with HSE06 functional of AlXY monolayers.

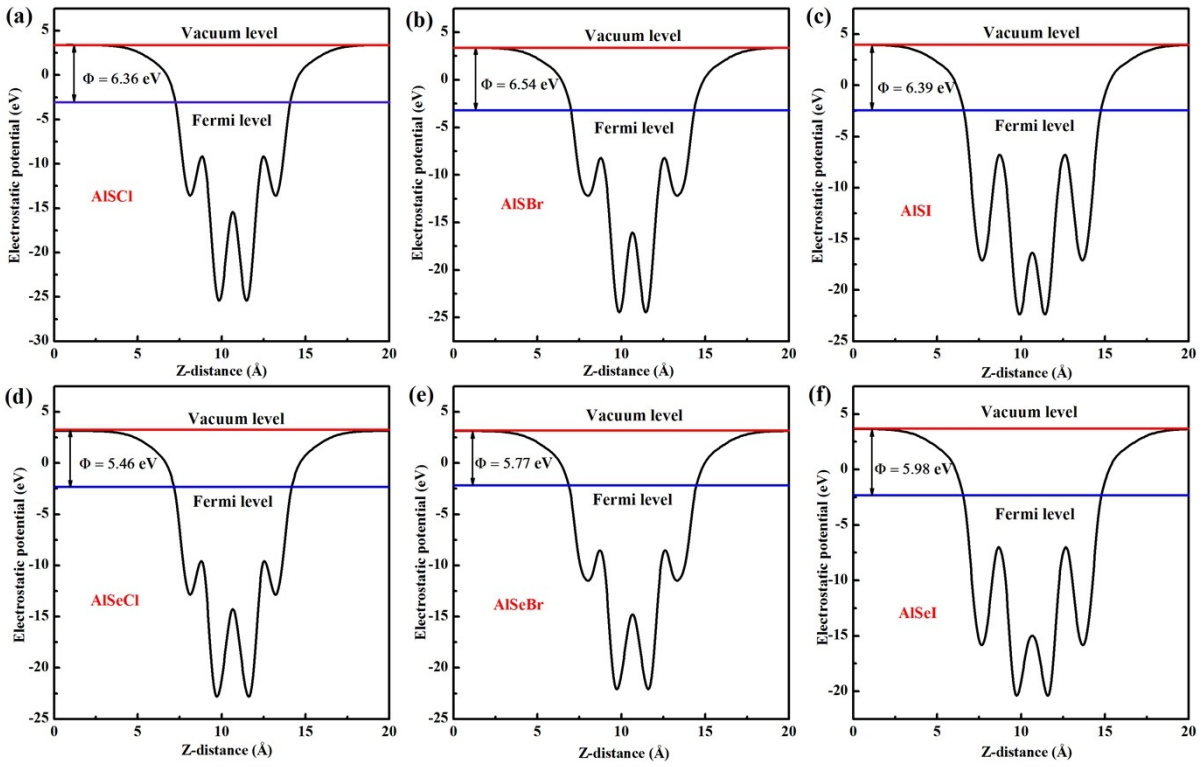

Fig. S6 (Color online) The calculated average electrostatic potential of AlXY monolayers under the HSE06 functional.