Supporting Information

Two-Dimensional Janus SeMoZAZ' (A=Si, Ge; Z= N, P, As; Z≠Z'): Multifunctional Properties for Electronics, Piezoelectric and Photocatalytic

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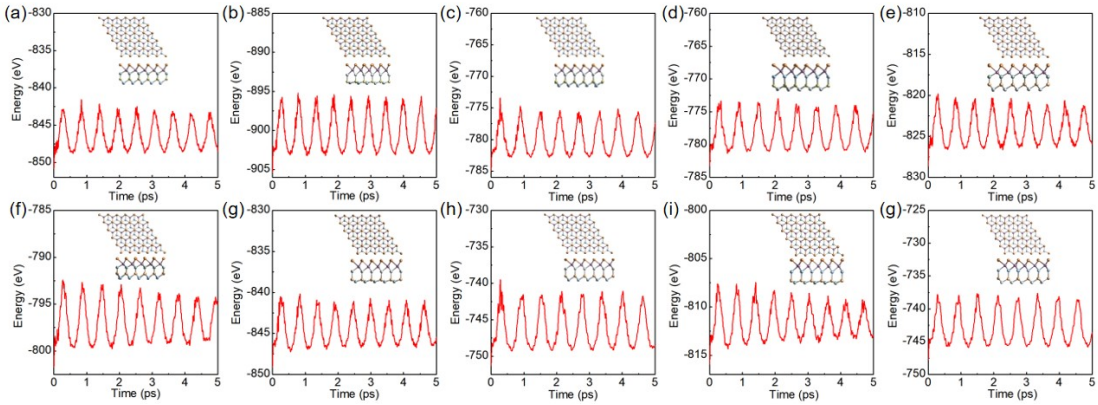

Fig S1 Variation of the energy as a function of time for the Janus (a) SeMoNSiAs, (b) SeMoPSiN, (c) SeMoPSiAs, (d) SeMoAsSiP, (e) SeMoNGeP, (f)SeMoNGeAs, (g) SeMoPGeN, (h) SeMoPGeAs, (i) SeMoAsGeN, and (j) SeMoAsGeP monolayers at 300K. The insets are the top and side views of the structure at the end of the AIMD simulation.

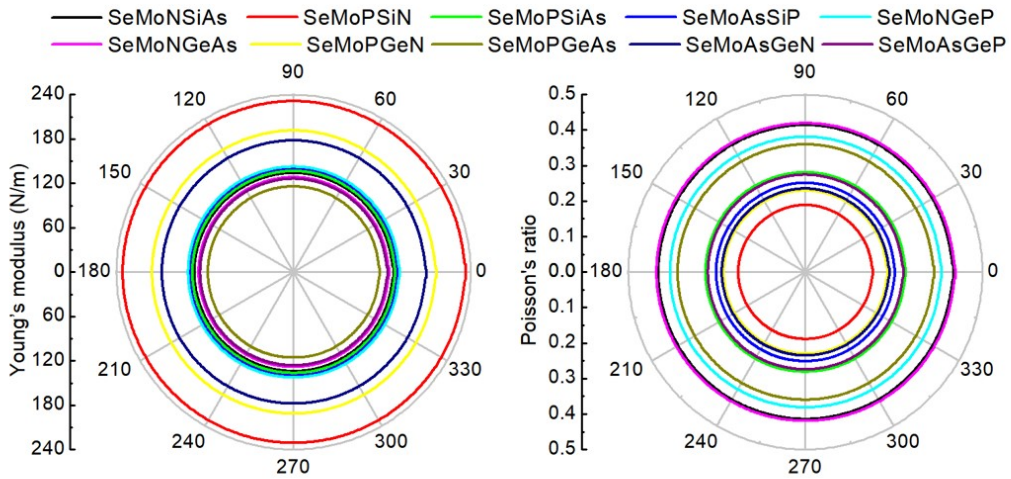

Fig S2 Young's modulus Y_{2D} and Poisson ratio ν of SeMoZAZ' (A=Si,Ge; Z,Z'=N, P, As; Z≠Z') monolayers.

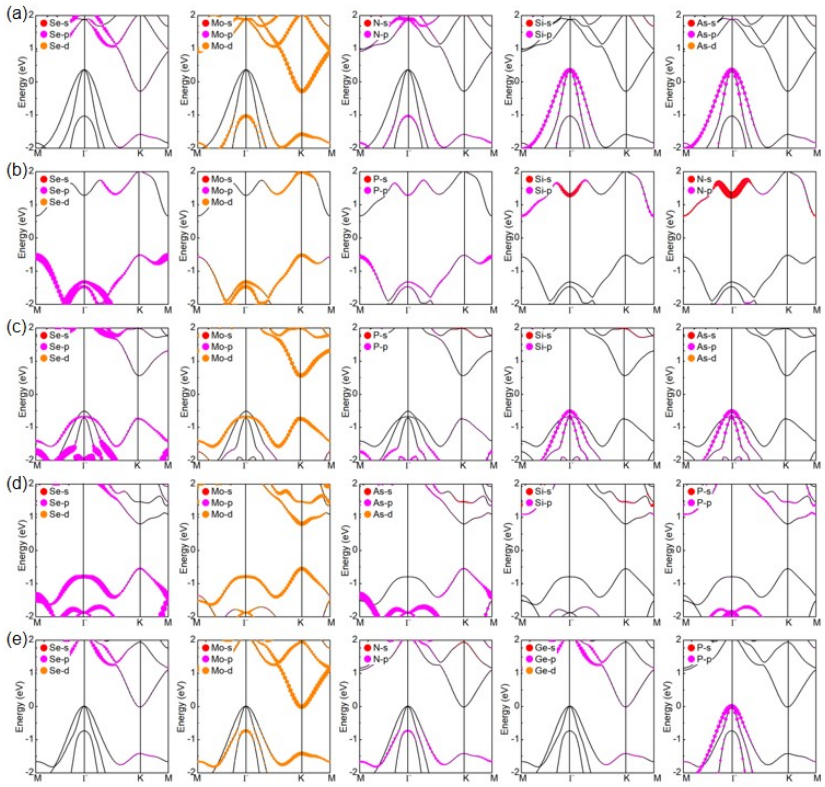

Fig S3 Projected band structure of 2D Janus (a) SeMoNSiAs, (b) SeMoPSiN, (c) SeMoPSiAs, (d) SeMoAsSiP, and (e) SeMoNGeP monolayers at equilibrium.

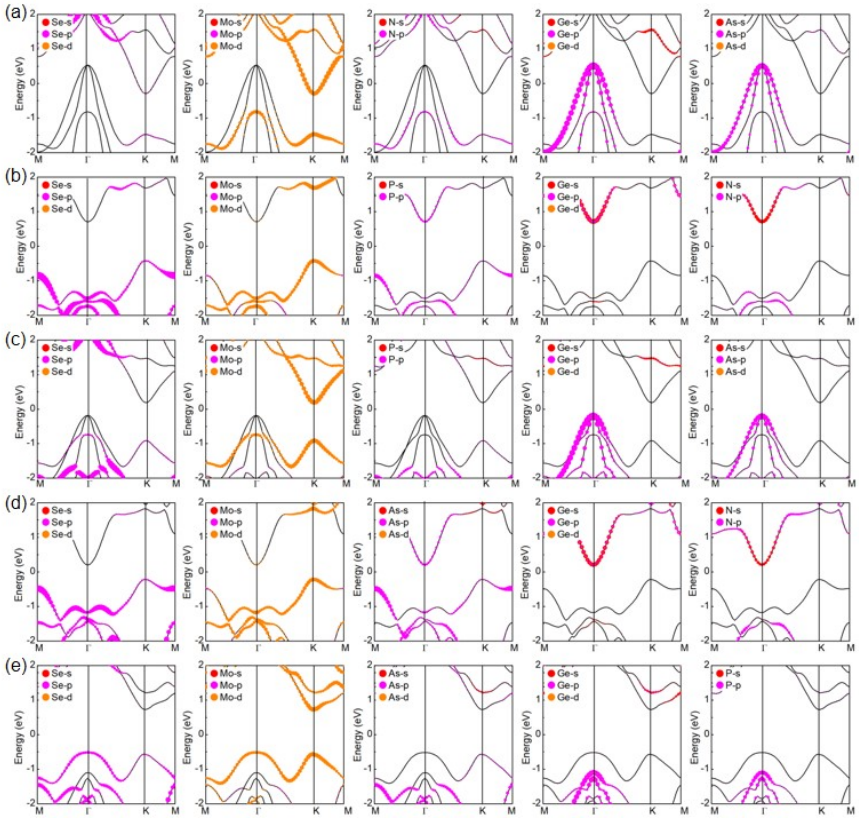

Fig S4 Projected band structure of 2D Janus (a)SeMoNGeAs, (b) SeMoPGeN, (c) SeMoPGeAs, (d) SeMoAsGeN, and (e) SeMoAsGeP monolayers at equilibrium.

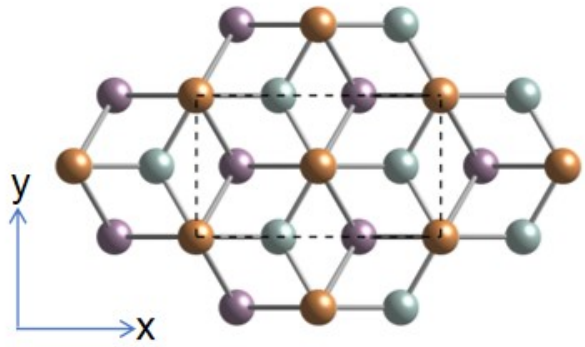

Fig S5 The calculation of piezoelectric stress coefficients and carrier mobility is carried out in rectangular cell in the black dotted box, the x and y directions also been indicate.

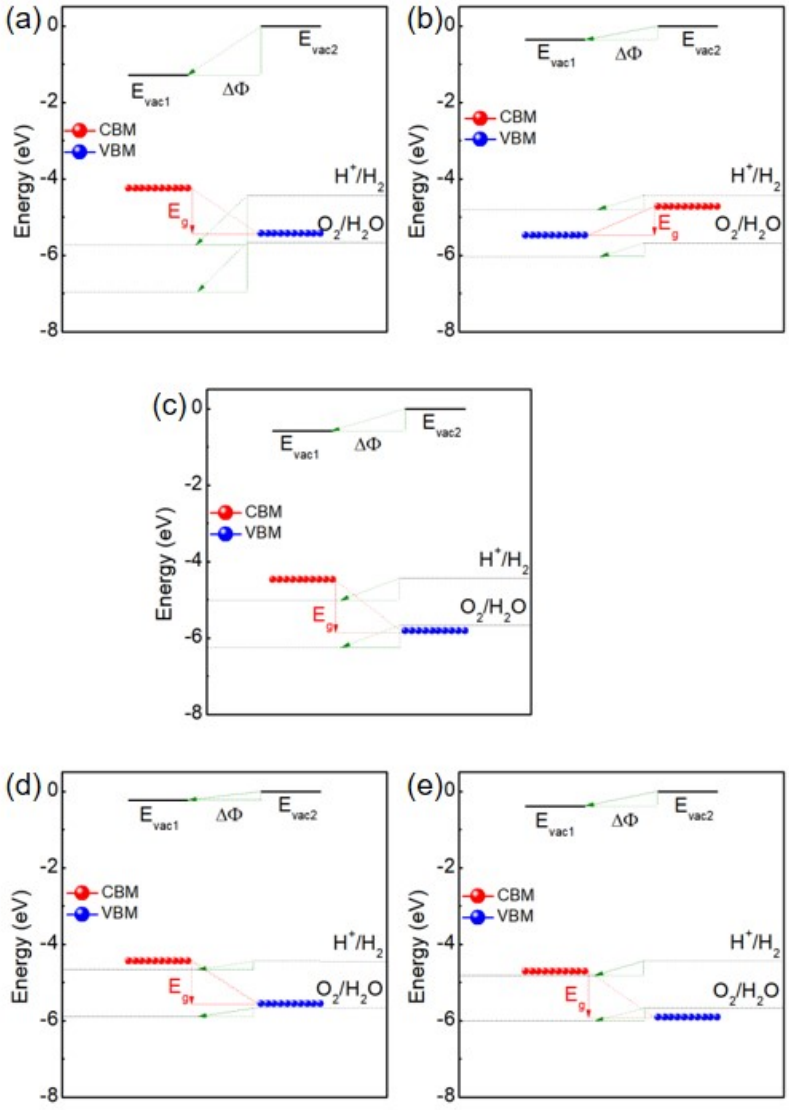

Fig S6 Band alignments with regard to the vacuum level of pristine (a) SeMoPSiN, (b) SeMoPSiAs (c)SeMoAsSiP, (d) SeMoPGeN and (e)SeMoAsGeP under the HSE06 functional.

The Solar-to-hydrogen (STH) Efficiency

The STH efficiency is evaluated using the methods proposed by Yang et al.[1] According to the reaction process, STH efficiency is defined as the product of the efficiency of light absorption (η_{abs}) and carrier utilization (η_{cu}).

$$\eta_{STH} = \eta_{abs} \times \eta_{cu} \tag{1}$$

The efficiency of light absorption is defined as:

$$\eta_{abs} = \frac{\int_{E_g}^{\infty} P(\mathrm{h}\omega) d(\mathrm{h}\omega)}{\int_{0}^{\infty} P(\mathrm{h}\omega) d(\mathrm{h}\omega)} \tag{2}$$

where $P(h\omega)$ are the AM1.5G solar energy flux at the photon energy $h\omega$ and E_g is the band gap of the materials. The denominator represents the total power density of the reference sunlight spectrum (AM1.5G) and the numerator gives the light power density absorbed by the photocatalyst.

The efficiency of carrier utilization (η_{cu}) is defined as:

$$\eta_{cu} = \frac{\Delta G_{H_2O} \int_{E}^{\infty} \frac{P(\mathrm{h}\omega)}{\mathrm{h}\omega} d(\mathrm{h}\omega)}{\int_{E_g}^{\infty} P(\mathrm{h}\omega) d(\mathrm{h}\omega)} \tag{3}$$

where ΔG_{H_2O} is the free energy of water splitting (1.23eV) and the rest of numerator represents the effective photocurrent density. Here, E represents the photon energy that can be actually utilized in the process of water splitting.

$$E = \begin{cases} E_g, \left(\chi(H_2) \geq 0.2, \chi(O_2) \geq 0.6\right) \\ E_g + 0.2 - \chi(H_2), \left(\chi(H_2) < 0.2, \chi(O_2) \geq 0.6\right) \\ E_g + 0.6 - \chi(O_2), \left(\chi(H_2) \geq 0.2, \chi(O_2) < 0.6\right) \\ E_g + 0.8 - \chi(H_2) - \chi(O_2), \left(\chi(H_2) < 0.2, \chi(O_2) < 0.6\right) \end{cases} \tag{4}$$

The intrinsic electric field does positive work for the electron-hole separation during the process of photocatalytic water splitting. Therefore, this part of work should be added into the total energy, and then the corrected STH efficiency of photocatalytic water splitting for 2D material with vertical intrinsic EF is calculated as:

$$\eta_{STH}^{'} = \eta_{STH} \times \frac{\int_0^{\infty} P(\mathrm{h}\omega) d(\mathrm{h}\omega)}{\int_0^{\infty} P(\mathrm{h}\omega) d(\mathrm{h}\omega) + \Delta V \int_{E_g}^{\infty} \frac{P(\mathrm{h}\omega)}{\mathrm{h}\omega} d(\mathrm{h}\omega)} \quad (5)$$

where ΔV is the vacuum level difference on the two respective surfaces.

1. C.-F. Fu, J. Sun, Q. Luo, X. Li, W. Hu and J. Yang, *Nano Lett.*, 2018, **18**, 6312-6317.