Supplementary information (SI) for

First-principles prediction of ferroelectric Janus Si$_2$XY (X/Y = S/Se/Te, X ≠ Y) monolayer with negative Poisson Ratios

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Table 1. Calculated in-plane elastic constants C$_{ij}$ (N m$^{-1}$) of Si$_2$SSe, Si$_2$STe and Si$_2$SeTe monolayer.

<table>
<thead>
<tr>
<th></th>
<th>C$_{11}$</th>
<th>C$_{12}$</th>
<th>C$_{22}$</th>
<th>C$_{66}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si$_2$SSe</td>
<td>89.753</td>
<td>-7.361</td>
<td>60.272</td>
<td>8.741</td>
</tr>
<tr>
<td>Si$_2$STe</td>
<td>76.360</td>
<td>-4.569</td>
<td>68.590</td>
<td>7.916</td>
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<tr>
<td>Si$_2$SeTe</td>
<td>67.235</td>
<td>-5.612</td>
<td>68.397</td>
<td>6.641</td>
</tr>
</tbody>
</table>

Fig.S1 Phonon dispersion of (a) Si$_2$STe and (b) Si$_2$SeTe monolayer, respectively.
Fig. S2 Total potential energy fluctuation and snapshots of atomic structure of (a) Si₂SSe, (b) Si₂STe and (c) Si₂SeTe monolayer at a temperature of 500 K from AIMD simulation under 0 ps and 10 ps.

Fig. S3 (a) The NPR of Janus Si₂XY monolayer as a function of the in-plane angle θ. The blue, pink and purple line represent Si₂SSe, Si₂STe and Si₂SeTe, respectively. (b) The strain-variation curve about the strain on a-axis with respect to the b direction. The evolution of local structures of the Janus Si₂XY monolayer in the a–c (c) and b–c (d) planes under uniaxial compression strain.
Fig. S4 Electronic band structures of (a) Si$_2$SSe, (b) Si$_2$STe and (c) Si$_2$SeTe monolayer with PBE calculation, respectively.

Fig. S5 The corresponding partial density of state (PDOS) of (a) Si$_2$STe and (b) Si$_2$SeTe monolayer, respectively.
Fig.S6 (a) Polarization P of monolayer Si$_2$SeTe as a function of normalized displacement along the $b$-axis. $P_s$ and $P_q$ represent the spontaneous and quanta polarization, respectively. (b) Energy curve with the $P_s$ of Si$_2$SeTe. Phase transition barrier $E_b$ and $P_s$ are marked. (c) Temperature dependence of the average relative displacement between ions in Si$_2$SeTe using MD simulations. The final atomic structures of Si$_2$SeTe near the phase transition temperature are depicted.

Fig.S7 The AIMD simulations of Si$_2$STe monolayer with the time step of 4 ps at the temperature of 1200 K. The white, green and orange balls denote Si, S and Te atoms, respectively.
Fig.S8 (a) Electronic band structure of monolayer Si₂SSe with PBE method. (b) The energy shifting and band-edge positions as a function of the (c) $a$-axis and $b$-axis strain along two transport directions of Si₂SSe monolayers. The solid lines indicate the fitting curves.
Fig. S9 (a) Electronic band structure of monolayer Si₂STe with PBE method. (b) The energy shifting and band-edge positions as a function of the (c) a-axis and b-axis strain along two transport directions of Si₂TSe monolayers. The solid lines indicate the fitting curves.
Fig. S10 (a) Electronic band structure of monolayer Si$_2$SeTe with PBE method. (b) The energy shifting and band-edge positions as a function of the (c) $a$-axis and $b$-axis strain along two transport directions of Si$_2$SeTe monolayers. The solid lines indicate the fitting curves.