Supporting Information

**Symmetry-breaking Induced Large Piezoelectricity in Janus Tellurene Materials**

Yu Chen ¹, Junyi Liu¹, Jiabing Yu¹, Yaguang Guo¹,², Qiang Sun ¹,² *

¹ Department of Materials Science and Engineering, Peking University, Beijing 100871, China

² Center for Applied Physics and Technology, Peking University, Beijing 100871, China

*To whom correspondence should be addressed. E-mail: sunqiang@pku.edu.cn
Figure S1. Phonon dispersion of the Janus Te$_2$Se multilayer structure (d). The high
symmetry k points are: $\Gamma$ (0, 0, 0), $K$ (-1/3, 2/3, 0), $M$ (0, 1/2, 0), $A$ (0, 0, 1/2), $H$ (-1/3,
2/3, 1/2), $L$ (0, 1/2, 1/2).

Table S1. Comparisons of clamped-ion elastic stiffness constants ($C_{ij}$) and the
clamped-ion piezoelectric coefficients ($e_{ij}/d_{ij}$) in unit of N/m, $10^{-10}$ C/m, and pm/V,
respectively.

<table>
<thead>
<tr>
<th>Materials</th>
<th>$C_{11}$</th>
<th>$C_{12}$</th>
<th>$e_{11}$</th>
<th>$e_{31}$</th>
<th>$d_{11}$</th>
<th>$d_{31}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Janus Te$_2$Se</td>
<td>57.92</td>
<td>15.33</td>
<td>3.994</td>
<td>0.146</td>
<td>9.378</td>
<td>0.199</td>
</tr>
<tr>
<td>$\alpha$-Tellurene</td>
<td>53.4</td>
<td>15.0</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>MoS$_2$</td>
<td>153</td>
<td>48</td>
<td>3.06</td>
<td>—</td>
<td>2.91</td>
<td>—</td>
</tr>
<tr>
<td>Janus In$_2$SSe*</td>
<td>88</td>
<td>27</td>
<td>7.94</td>
<td>0.07</td>
<td>13.06</td>
<td>0.11</td>
</tr>
</tbody>
</table>

Table S2. Relative energy per unit cell ($\Delta E$), lattice parameters ($a/c$) and the unit-cell
volume ($V$) of the unit cell of Janus Te$_2$Se multilayers with the structures shown in
Figure 6. The units of $\Delta E$, $a/c$, and $V$ are meV, Å, and (Å$^3$), respectively.

<table>
<thead>
<tr>
<th>Structure</th>
<th>$\Delta E$</th>
<th>$a$</th>
<th>$c$</th>
<th>$V$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>0</td>
<td>4.157</td>
<td>12.115</td>
<td>181.33</td>
</tr>
<tr>
<td>(b)</td>
<td>208</td>
<td>4.099</td>
<td>13.573</td>
<td>197.52</td>
</tr>
<tr>
<td>(c)</td>
<td>315</td>
<td>4.091</td>
<td>14.267</td>
<td>206.76</td>
</tr>
<tr>
<td>(d)</td>
<td>47</td>
<td>4.130</td>
<td>12.504</td>
<td>184.71</td>
</tr>
<tr>
<td>(e)</td>
<td>68</td>
<td>4.118</td>
<td>12.688</td>
<td>186.33</td>
</tr>
</tbody>
</table>
Table S3. The clamped-ion elastic stiffness coefficients ($C_{ij}$) and the clamped-ion piezoelectric coefficients ($e_{ij}/d_{ij}$) of the Janus Te$_2$Se multilayer with five structures in Figure 6. The units of $C_{ij}$, $e_{ij}$ and $d_{ij}$ are GPa, $10^{-10}$ C/m and pm/V, respectively.

<table>
<thead>
<tr>
<th>Structure</th>
<th>$C_{11}$</th>
<th>$C_{12}$</th>
<th>$C_{13}$</th>
<th>$C_{33}$</th>
<th>$e_{11}$</th>
<th>$e_{31}$</th>
<th>$e_{33}$</th>
<th>$d_{11}$</th>
<th>$d_{31}$</th>
<th>$d_{33}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>88.08</td>
<td>22.78</td>
<td>46.23</td>
<td>89.10</td>
<td>0.755</td>
<td>0.170</td>
<td>-0.428</td>
<td>11.562</td>
<td>6.232</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>11.269</td>
</tr>
<tr>
<td>(b)</td>
<td>81.53</td>
<td>22.85</td>
<td>37.22</td>
<td>83.92</td>
<td>0.601</td>
<td>0.098</td>
<td>-0.177</td>
<td>10.248</td>
<td>2.473</td>
<td>-4.308</td>
</tr>
<tr>
<td>(c)</td>
<td>78.90</td>
<td>21.59</td>
<td>34.02</td>
<td>80.52</td>
<td>0</td>
<td>0.089</td>
<td>-0.157</td>
<td>0</td>
<td>2.163</td>
<td>-3.777</td>
</tr>
<tr>
<td>(d)</td>
<td>82.95</td>
<td>26.59</td>
<td>44.65</td>
<td>85.20</td>
<td>0.076</td>
<td>-0.178</td>
<td>0</td>
<td>2.709</td>
<td>-4.934</td>
<td></td>
</tr>
<tr>
<td>(e)</td>
<td>85.20</td>
<td>23.19</td>
<td>42.43</td>
<td>84.45</td>
<td>0.143</td>
<td>-0.322</td>
<td>0</td>
<td>4.637</td>
<td>-8.469</td>
<td></td>
</tr>
</tbody>
</table>

Reference
