Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.
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# Electronic Supplementary Information

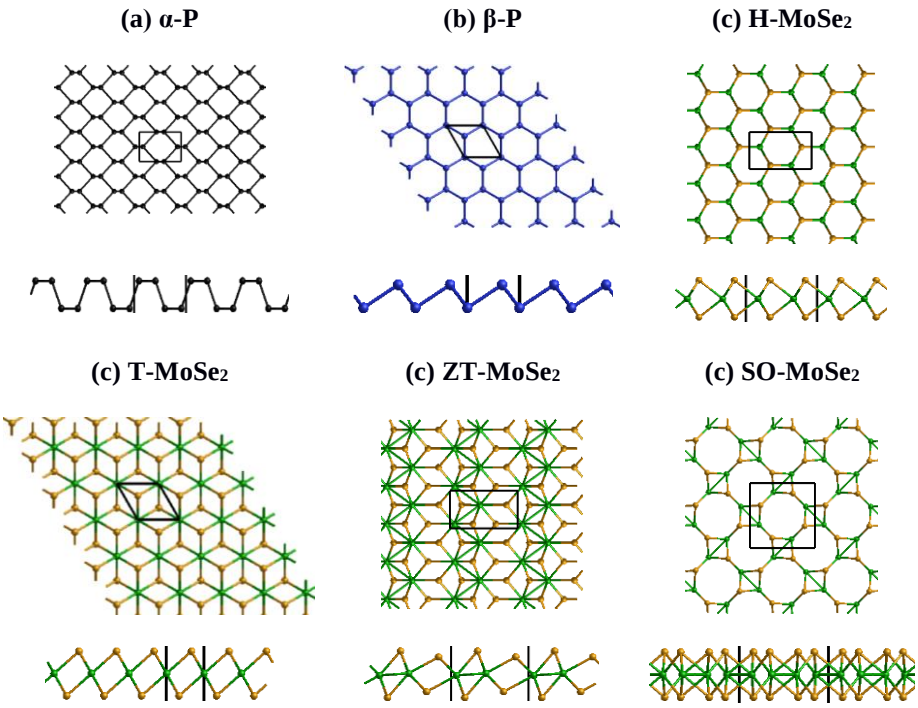


**FIGURE S1.** Top and side view of monolayer. Black and blue balls in (a) and (b) indicate P atoms while green and yellow balls in (c), (d) and (f) indicate Mo and Se atoms, respectively.

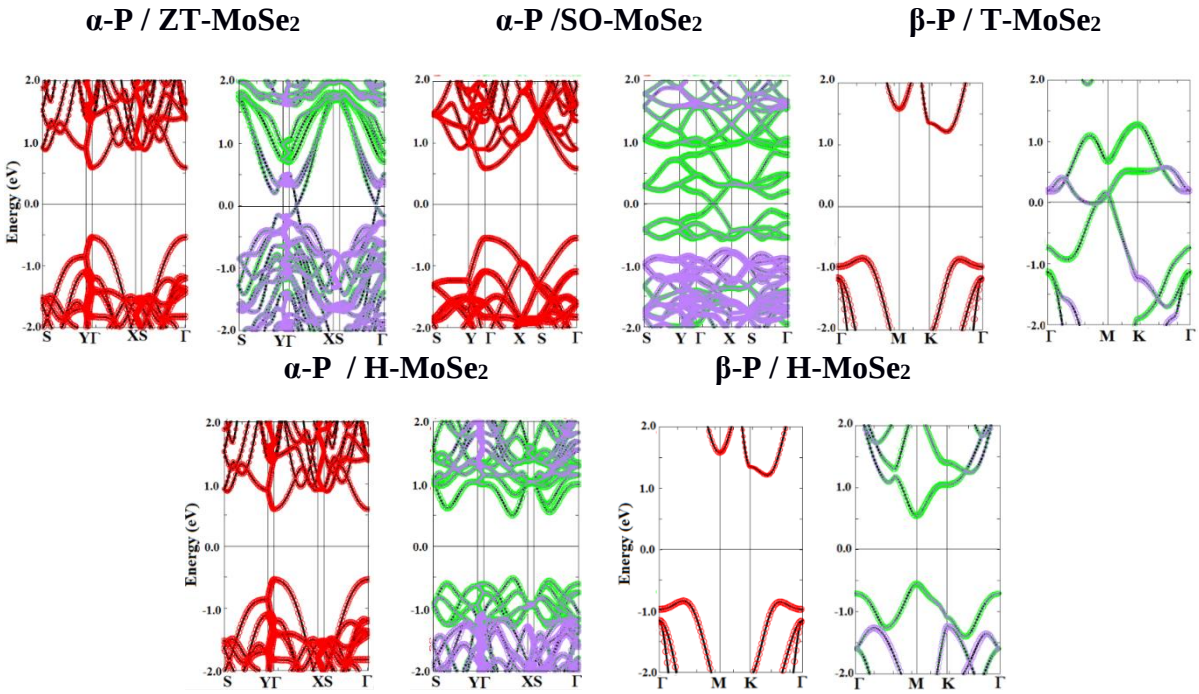


**FIGURE S2.** The bandstructure of isolated monolayers within the heterostructures supercell. Red color indicate bands corresponding to phosphorene while green and purple color in the other bandstructure ($MoSe_2$) depicts Mo and Se contributions respectively.

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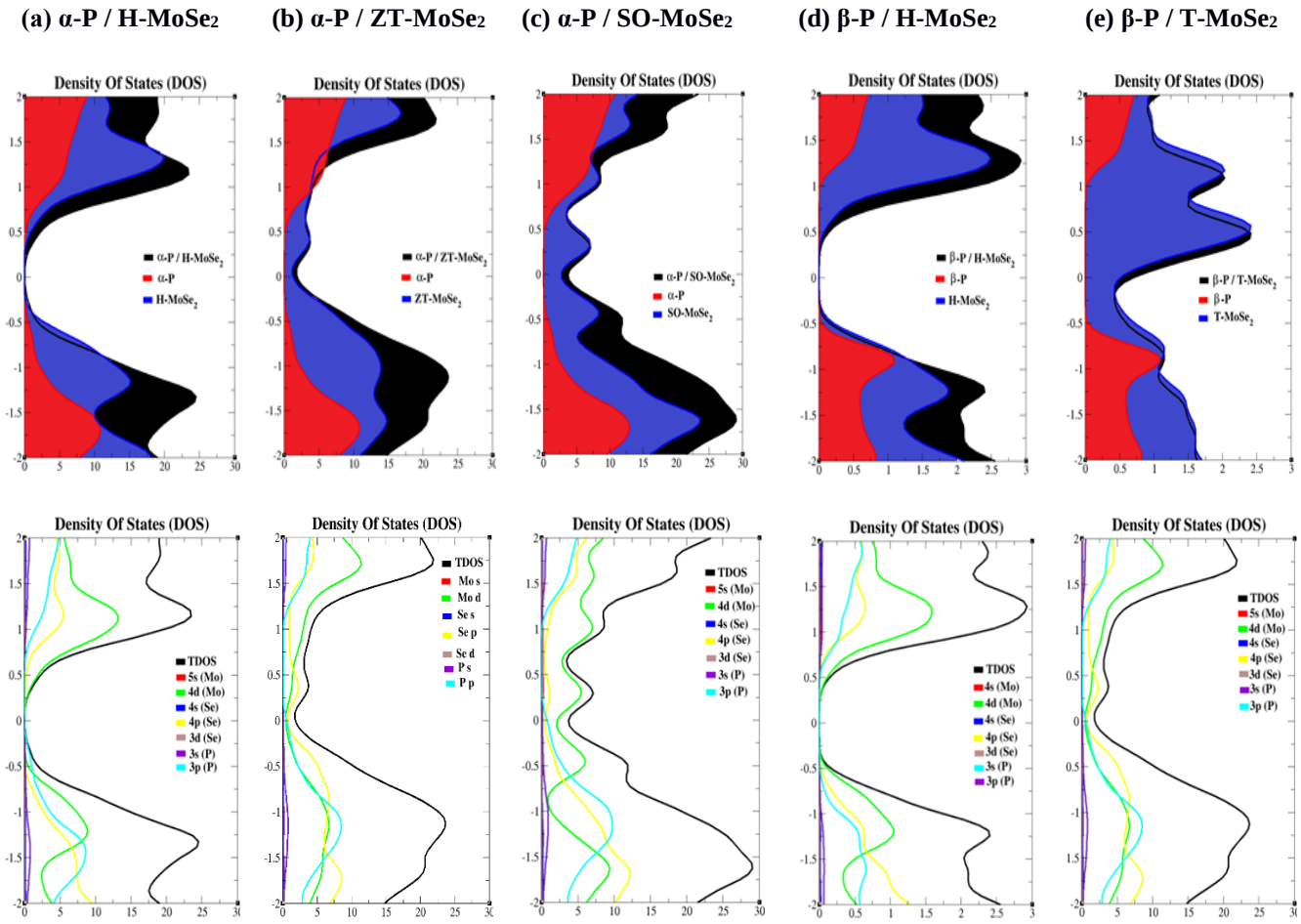


**FIGURE S3.** Total density of states along with the orbital resolved density of states of the heterostructures (a) α-P / H-$MoSe_2$, (b) α-P / ZT-$MoSe_2$, (c) α-P / SO-$MoSe_2$, (d) β-P / H-$MoSe_2$ and (e) β-P / T-$MoSe_2$.

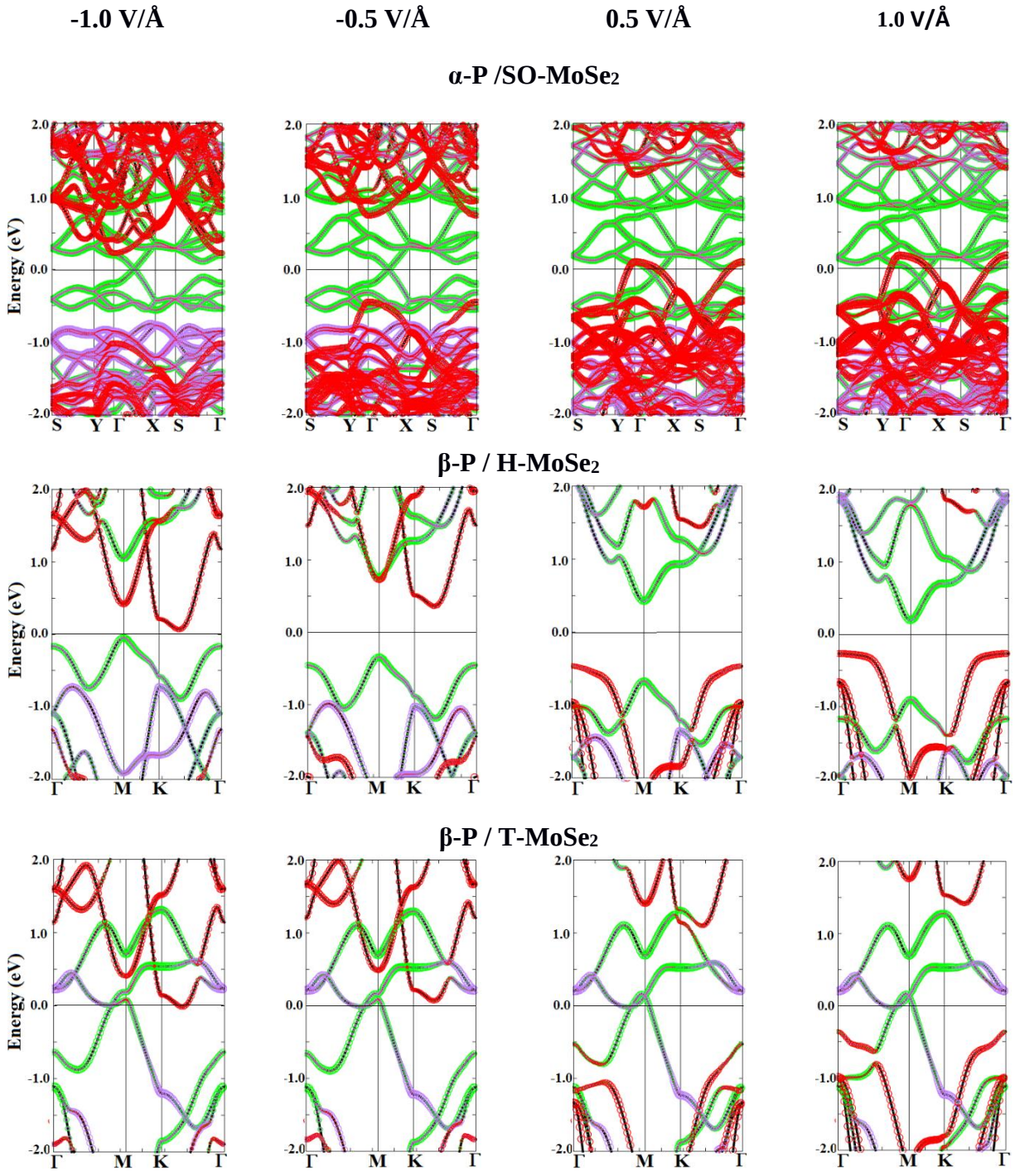


**FIGURE S4.** Evolution of band structure on the function of external electric field. Red, green and purple color in the band structure indicates the contribution of 3p orbitals of P, 4d orbitals of Mo and 4p orbitals of Se respectively.

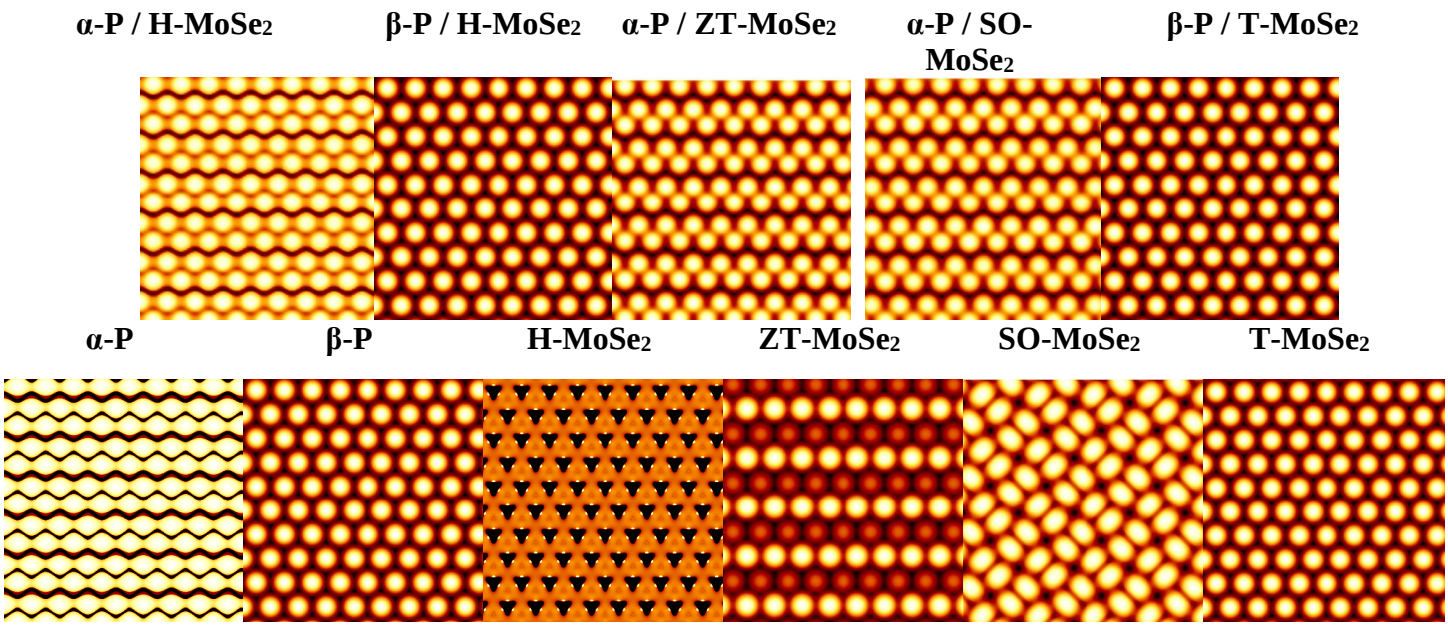


**FIGURE S5.** Simulated STM images of dimension 30 Å × 30 Å for various heterostructures compared with the monolayers. The isosurface is taken at 1.0 $\times 10^{-6}$ e/Å$^3$.The bias voltage for α-P based heterostructures is - 0.5 V and for β-P based heterostructures it is -1.5 V.

# Electronic Supplementary Information

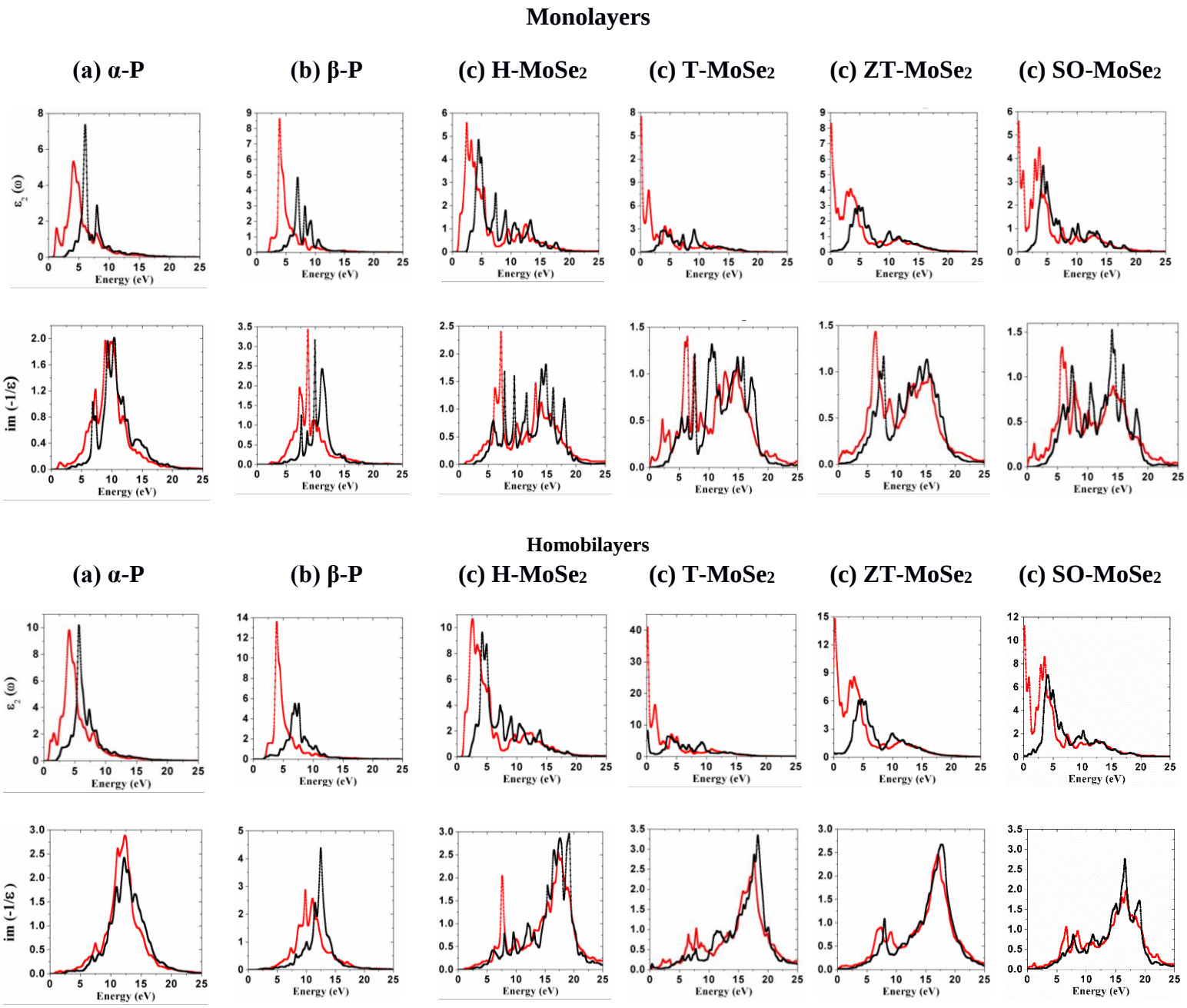


**FIGURE S6.** Imaginary part of the dielectric constant ($\varepsilon_2(\omega)$) and electron energy loss spectra (im (-1/ε)) of monolayers and homobilayers of (a) α-P, (b) β-P, (c) H-$MoSe_2$ (hexagonal unitcell), (d) T-$MoSe_2$, (e) ZT-$MoSe_2$ and (f) SO-$MoSe_2$. Red color is for lateral polarization while black is for vertical polarization.

1. **The structural information about α-P / SO-$MoSe_2$**

Total Energy of α-P / SO-$MoSe_2$ is -10184.5522 eV

| | |
|---|---|
| _cell_length_a | 6.8273 |
| _cell_length_b | 14.5061 |
| _cell_length_c | 24.7740 |
| _cell_angle_alpha | 90.00 |
| _cell_angle_beta | 90.00 |
| _cell_angle_gamma | 90.00 |

Atomic Coordinates Format: Fractional

| | | | |
|---|---|---|---|
| Mo | 0.3614 | 0.4307 | 0.1308 |
| Se | 0.6602 | 0.4199 | 0.0629 |
| Se | 0.6602 | 0.4199 | 0.1986 |
| Se | 0.1602 | 0.3301 | 0.0629 |
| Se | 0.1602 | 0.3301 | 0.1986 |
| Mo | 0.1386 | 0.1807 | 0.1308 |
| Mo | 0.8614 | 0.3193 | 0.1308 |
| Se | 0.8398 | 0.1699 | 0.0629 |
| Se | 0.8398 | 0.1699 | 0.1986 |
| Se | 0.3398 | 0.0801 | 0.0629 |
| Se | 0.3398 | 0.0801 | 0.1986 |
| Mo | 0.6386 | 0.0693 | 0.1308 |
| Mo | 0.3614 | 0.9307 | 0.1308 |
| Se | 0.6602 | 0.9199 | 0.0629 |
| Se | 0.6602 | 0.9199 | 0.1986 |
| Se | 0.1602 | 0.8301 | 0.0629 |
| Se | 0.1602 | 0.8301 | 0.1986 |
| Mo | 0.1386 | 0.6807 | 0.1308 |
| Mo | 0.8614 | 0.8193 | 0.1308 |
| Se | 0.8398 | 0.6699 | 0.0629 |
| Se | 0.8398 | 0.6699 | 0.1986 |
| Se | 0.3398 | 0.5801 | 0.0629 |
| Se | 0.3398 | 0.5801 | 0.1986 |
| Mo | 0.6386 | 0.5693 | 0.1308 |
| P | 0.3750 | 0.0303 | 0.4363 |
| P | 0.1250 | 0.1364 | 0.4363 |
| P | 0.1250 | 0.1969 | 0.3480 |
| P | 0.3750 | 0.3031 | 0.3480 |
| P | 0.8750 | 0.6969 | 0.4363 |
| P | 0.6250 | 0.8031 | 0.4363 |
| P | 0.6250 | 0.8636 | 0.3480 |
| P | 0.8750 | 0.9698 | 0.3480 |
| P | 0.3750 | 0.6969 | 0.4363 |
| P | 0.1250 | 0.8031 | 0.4363 |
| P | 0.1250 | 0.8636 | 0.3480 |
| P | 0.3750 | 0.9698 | 0.3480 |
| P | 0.8750 | 0.3636 | 0.4363 |

| | | | |
|---|---|---|---|
| P | 0.6250 | 0.4697 | 0.4363 |
| P | 0.6250 | 0.5302 | 0.3480 |
| P | 0.8750 | 0.6364 | 0.3480 |
| P | 0.3750 | 0.3636 | 0.4363 |
| P | 0.1250 | 0.4697 | 0.4363 |
| P | 0.1250 | 0.5302 | 0.3480 |
| P | 0.3750 | 0.6364 | 0.3480 |
| P | 0.8750 | 0.0303 | 0.4363 |
| P | 0.6250 | 0.1364 | 0.4363 |
| P | 0.6250 | 0.1969 | 0.3480 |
| P | 0.8750 | 0.3031 | 0.3480 |

## 2. The structural information about β-P / H-$MoSe_2$

Total Energy of β-P / H-$MoSe_2$ is -1094.3936 eV

| | |
|---|---|
| _cell_length_a | 3.4135 |
| _cell_length_b | 3.4135 |
| _cell_length_c | 24.6806 |
| _cell_angle_alpha | 90.00 |
| _cell_angle_beta | 90.00 |
| _cell_angle_gamma | 120.00 |

Atomic Coordinates Format: Fractional

| | | | |
|---|---|---|---|
| Mo | 0.2710 | 0.6172 | 0.1301 |
| Se | 0.6044 | 0.2838 | 0.0618 |
| Se | 0.6045 | 0.2841 | 0.1984 |
| P | 0.9989 | 0.0011 | 0.3524 |
| P | 0.3323 | 0.6678 | 0.4038 |