Journal Name

ARTICLE TYPE

Cite this: DOI: 10.1039/xxxxxxxxxx

Layering Effects on Low Frequency Modes in *n*-layered MX_2 Transition Metal Dichalcogenides - Electronic Supplementary Information

Antonio Cammarata,*[a] and Tomas Polcar[b]

Received Date
Accepted Date

DOI: 10.1039/xxxxxxxxxx

www.rsc.org/journalname

1 Optimized Structures of the MX-*n*L Systems

In the following subsections, we report the optimized structure of each MX_2 bulk system we used to build the corresponding MX-*n*L model geometries. Structures are reported in "cif" format.

1.1 MoS_2

```
_symmetry_space_group_name_H-M "P 63/m 2/m 2/c"
_symmetry_Int_Tables_number 194

_cell_length_a    3.19183
_cell_length_b    3.19183
_cell_length_c    12.41278
_cell_angle_alpha 90.00000
_cell_angle_beta  90.00000
_cell_angle_gamma 120.00000

loop_
_space_group_symop_id
_space_group_symop_operation_xyz
1 x,y,z
2 x-y,x,z+1/2
3 -y,x-y,z
4 -x,-y,z+1/2
5 -x+y,-x,z
6 y,-x+y,z+1/2
7 x-y,-y,-z
8 x,x-y,-z+1/2
9 y,x,-z
10 -x+y,y,-z+1/2
11 -x,-x+y,-z
12 -y,-x,-z+1/2
13 -x,-y,-z
14 -x+y,-x,-z+1/2
15 y,-x+y,-z
16 x,y,-z+1/2
17 x-y,x,-z
18 -y,x-y,-z+1/2
19 -x+y,y,z
20 -x,-x+y,z+1/2
21 -y,-x,z
22 x-y,-y,z+1/2
23 x,x-y,z
24 y,x,z+1/2

loop_
_atom_site_label
_atom_site_type_symbol
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_label
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
Mo1 Mo   2 d 0.33333 0.66667 0.75000 1.00000
S1 S   4 f 0.33333 0.66667 0.12474 1.00000
```

1.2 $MoSe_2$

```
_symmetry_space_group_name_H-M "P 63/m 2/m 2/c"
_symmetry_Int_Tables_number 194

_cell_length_a    3.32053
_cell_length_b    3.32053
_cell_length_c    13.05794
_cell_angle_alpha 90.00000
```

[a] Department of Control Engineering, Czech Technical University in Prague, Technicka 2, 16627 Prague 6, Czech Republic. Fax: +420 224 91 8646; Tel: +420 224 35 7598; E-mail: cammaant@fel.cvut.cz
[b] nCATS, FEE, University of Southampton, SO17 1BJ Southampton, United Kingdom.

```
_cell_angle_beta  90.00000
_cell_angle_gamma 120.00000

loop_
_space_group_symop_id
_space_group_symop_operation_xyz
1 x,y,z
2 x-y,x,z+1/2
3 -y,x-y,z
4 -x,-y,z+1/2
5 -x+y,-x,z
6 y,-x+y,z+1/2
7 x-y,-y,-z
8 x,x-y,-z+1/2
9 y,x,-z
10 -x+y,y,-z+1/2
11 -x,-x+y,-z
12 -y,-x,-z+1/2
13 -x,-y,-z
14 -x+y,-x,-z+1/2
15 y,-x+y,-z
16 x,y,-z+1/2
17 x-y,x,-z
18 -y,x-y,-z+1/2
19 -x+y,y,z
20 -x,-x+y,z+1/2
21 -y,-x,z
22 x-y,-y,z+1/2
23 x,x-y,z
24 y,x,z+1/2

loop_
_atom_site_label
_atom_site_type_symbol
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_label
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
Mo1 Mo   2 d 0.33333 0.66667 0.75000 1.00000
Se1 Se   4 f 0.33333 0.66667 0.12245 1.00000
```

1.3 $MoTe_2$

```
_symmetry_space_group_name_H-M "P 63/m 2/m 2/c"
_symmetry_Int_Tables_number 194

_cell_length_a    3.53284
_cell_length_b    3.53284
_cell_length_c    14.01184
_cell_angle_alpha 90.00000
_cell_angle_beta  90.00000
_cell_angle_gamma 120.00000

loop_
_space_group_symop_id
_space_group_symop_operation_xyz
1 x,y,z
2 x-y,x,z+1/2
3 -y,x-y,z
4 -x,-y,z+1/2
5 -x+y,-x,z
6 y,-x+y,z+1/2
7 x-y,-y,-z
8 x,x-y,-z+1/2
9 y,x,-z
10 -x+y,y,-z+1/2
11 -x,-x+y,-z
12 -y,-x,-z+1/2
13 -x,-y,-z
14 -x+y,-x,-z+1/2
15 y,-x+y,-z
16 x,y,-z+1/2
17 x-y,x,-z
18 -y,x-y,-z+1/2
19 -x+y,y,z
20 -x,-x+y,z+1/2
21 -y,-x,z
22 x-y,-y,z+1/2
23 x,x-y,z
24 y,x,z+1/2

loop_
_atom_site_label
_atom_site_type_symbol
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_label
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
Mo1 Mo   2 d 0.33333 0.66667 0.75000 1.00000
Te1 Te   4 f 0.33333 0.66667 0.12077 1.00000
```

1.4 WS_2

```
_symmetry_space_group_name_H-M "P 63/m 2/m 2/c"
_symmetry_Int_Tables_number 194

_cell_length_a    3.17781
_cell_length_b    3.17781
_cell_length_c    12.41916
_cell_angle_alpha 90.00000
_cell_angle_beta  90.00000
_cell_angle_gamma 120.00000

loop_
_space_group_symop_id
_space_group_symop_operation_xyz
1 x,y,z
2 x-y,x,z+1/2
```

```
3 -y,x-y,z
4 -x,-y,z+1/2
5 -x+y,-x,z
6 y,-x+y,z+1/2
7 x-y,-y,-z
8 x,x-y,-z+1/2
9 y,x,-z
10 -x+y,y,-z+1/2
11 -x,-x+y,-z
12 -y,-x,-z+1/2
13 -x,-y,-z
14 -x+y,-x,-z+1/2
15 y,-x+y,-z
16 x,y,-z+1/2
17 x-y,x,-z
18 -y,x-y,-z+1/2
19 -x+y,y,z
20 -x,-x+y,z+1/2
21 -y,-x,z
22 x-y,-y,z+1/2
23 x,x-y,z
24 y,x,z+1/2

loop_
_atom_site_label
_atom_site_type_symbol
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_label
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
W1 W   2 d 0.33333 0.66667 0.75000 1.00000
S1 S   4 f 0.33333 0.66667 0.12377 1.00000
```

1.5 WSe_2

```
_symmetry_space_group_name_H-M "P 63/m 2/m 2/c"
_symmetry_Int_Tables_number 194

_cell_length_a    3.32124
_cell_length_b    3.32124
_cell_length_c    13.11919
_cell_angle_alpha 90.00000
_cell_angle_beta  90.00000
_cell_angle_gamma 120.00000

loop_
_space_group_symop_id
_space_group_symop_operation_xyz
1 x,y,z
2 x-y,x,z+1/2
3 -y,x-y,z
4 -x,-y,z+1/2
5 -x+y,-x,z
6 y,-x+y,z+1/2
7 x-y,-y,-z
8 x,x-y,-z+1/2
9 y,x,-z
10 -x+y,y,-z+1/2
11 -x,-x+y,-z
12 -y,-x,-z+1/2
13 -x,-y,-z
14 -x+y,-x,-z+1/2
15 y,-x+y,-z
16 x,y,-z+1/2
17 x-y,x,-z
18 -y,x-y,-z+1/2
19 -x+y,y,z
20 -x,-x+y,z+1/2
21 -y,-x,z
22 x-y,-y,z+1/2
23 x,x-y,z
24 y,x,z+1/2

loop_
_atom_site_label
_atom_site_type_symbol
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_label
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
W1 W   2 c 0.33333 0.66667 0.25000 1.00000
Se1 Se   4 f 0.33333 0.66667 0.62242 1.00000
```

1.6 WTe_2

```
_symmetry_space_group_name_H-M "P 63/m 2/m 2/c"
_symmetry_Int_Tables_number 194

_cell_length_a    3.52273
_cell_length_b    3.52273
_cell_length_c    14.44008
_cell_angle_alpha 90.00000
_cell_angle_beta  90.00000
_cell_angle_gamma 120.00000

loop_
_space_group_symop_id
_space_group_symop_operation_xyz
1 x,y,z
2 x-y,x,z+1/2
3 -y,x-y,z
4 -x,-y,z+1/2
5 -x+y,-x,z
6 y,-x+y,z+1/2
7 x-y,-y,-z
8 x,x-y,-z+1/2
9 y,x,-z
10 -x+y,y,-z+1/2
```

```
11 -x,-x+y,-z
12 -y,-x,-z+1/2
13 -x,-y,-z
14 -x+y,-x,-z+1/2
15 y,-x+y,-z
16 x,y,-z+1/2
17 x-y,x,-z
18 -y,x-y,-z+1/2
19 -x+y,y,z
20 -x,-x+y,z+1/2
21 -y,-x,z
22 x-y,-y,z+1/2
23 x,x-y,z
24 y,x,z+1/2

loop_
_atom_site_label
_atom_site_type_symbol
_atom_site_symmetry_multiplicity
_atom_site_Wyckoff_label
_atom_site_fract_x
_atom_site_fract_y
_atom_site_fract_z
_atom_site_occupancy
W1 W   2 d 0.33333 0.66667 0.75000 1.00000
Te1 Te   4 f 0.33333 0.66667 0.12335 1.00000
```

2 "In-phase" and "Out-of-phase" Layer Shifts

The mode displacements, individuated in bi-layer ($n = 2$) systems, become more and more complex at increasing number of layers n. In 3- and 4-layer systems, both sliding and variable ID modes can be realized by in-phase or out-of-phase layer shifts (Figure 1). Considering two adjacent layers, we can define: *i*) "in-phase" layer shift, consisting of parallel layer displacements towards the same direction, one layer moving at a velocity different than that of its adjacent layer(s); *ii*) "out-of-phase" layer shift, consisting of antiparallel layer displacements (then towards opposite directions).

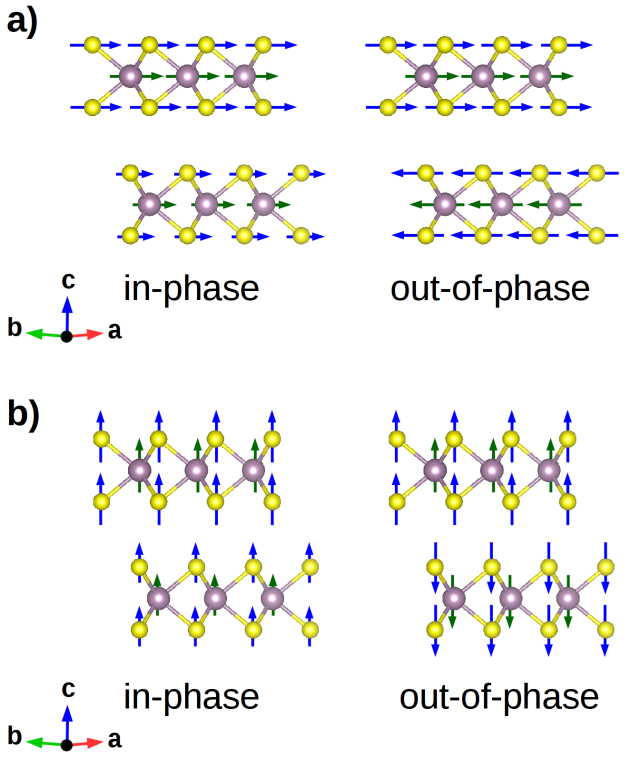

Fig. 1 At $n = 3,4$, (a) sliding and (b) variable ID modes can be realized by in-phase or out-of-phase layer shifts. In in-phase shifts, adjacent layers displace at different velocities, represented by arrows with different length.