# Supplementary Information

## Superconductivity in o-MAX phases

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**Table1S.** Lattice parameters, volume and formation energy of the hexagonal $M'_2MAlC_2$.

| Material | Formation Energy (eV/atom) | *a(A)* | *c(A)* | Volume($A^3$) |
|---|---|---|---|---|
| **$Mo_2HfAlC_2$** | -0.476 | 3.07 | 19.24 | 156.39 |
| **$Mo_2NbAlC_2$** | -0.313 | 3.07 | 18.75 | 153.08 |
| **$Mo_2ScAlC_2$** | -0.387 | 3.05 | 19.08 | 153.88 |
| **$Mo_2TaAlC_2$** | -0.342 | 3.06 | 18.74 | 152.45 |
| **$Mo_2TiAlC_2$** | -0.470 | 2.99 | 18.77 | 146.32 |
| **$Mo_2VAlC_2$** | -0.287 | 2.97 | 18.57 | 142.61 |
| **$Mo_2YAlC_2$** | -0.214 | 3.14 | 19.42 | 166.16 |
| **$Mo_2ZrAlC_2$** | -0.416 | 3.10 | 18.99 | 158.30 |
| **Material** | **Formation Energy (eV/atom)** | ***a(A)*** | ***c(A)*** | **volume($A^3$)** |
| **$Nb_2HfAlC_2$** | -0.710 | 3.16 | 19.22 | 167.03 |
| **$Nb_2ScAlC_2$** | -0.596 | 3.18 | 19.21 | 168.43 |
| **$Nb_2TaAlC_2$** | -0.617 | 3.11 | 19.23 | 161.24 |
| **$Nb_2TiAlC_2$** | -0.673 | 3.10 | 18.89 | 157.88 |
| **$Nb_2VAlC_2$** | -0.512 | 3.04 | 19.20 | 153.88 |
| **$Nb_2YAlC_2$** | -0.461 | 3.25 | 19.75 | 181.39 |
| **$Nb_2ZrAlC_2$** | -0.663 | 3.17 | 19.69 | 171.71 |
| **Material** | **Formation Energy (eV/atom)** | ***a(A)*** | ***c(A)*** | **volume($A^3$)** |
| **$W_2HfAlC_2$** | -0.375 | 3.05 | 19.24 | 154.92 |
| **$W_2NbAlC_2$** | -0.176 | 3.04 | 19.19 | 154.77 |
| **$W_2ScAlC_2$** | -0.328 | 3.02 | 19.29 | 152.74 |
| **$W_2TaAlC_2$** | -0.201 | 3.05 | 18.80 | 151.96 |
| **$W_2TiAlC_2$** | -0.378 | 2.98 | 18.88 | 145.97 |
| **$W_2VAlC_2$** | -0.167 | 2.96 | 18.69 | 142.45 |
| **$W_2YAlC_2$** | -0.137 | 3.12 | 19.83 | 167.99 |
| **$W_2ZrAlC_2$** | -0.309 | 3.07 | 19.47 | 157.49 |

**Table 2S.** ICOHP and bond length of different bonding in $M'_2MAlC_2$ structures.

| $W_2TaAlC_2$ | | | $W_2VAlC_2$ | | | $W_2NbAlC_2$ | | | $Mo_2NbAlC_2$ | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) |
| W-W | 3.05 | -1.08 | W-W | 2.96 | -1.31 | W-W | 3.04 | -1.11 | Mo-Mo | 3.07 | -0.80 |
| W-Ta | 3.07 | -1.13 | W-V | 2.99 | -0.86 | W-Nb | 3.10 | -0.96 | Mo-Nb | 3.06 | -0.85 |
| W-Al | 2.80 | -2.22 | W-Al | 2.80 | -2.19 | W-Al | 2.84 | -2.12 | Mo-Al | 2.81 | -1.87 |
| W-C | 2.12 | -4.19 | W-C | 2.11 | -4.49 | W-C | 2.12 | -4.23 | Mo-C | 2.11 | -4.02 |
| Ta-C | 2.20 | -3.93 | V-C | 2.10 | 3.03 | Nb-C | 2.22 | -3.58 | Nb-C | 2.22 | -3.61 |
| Ta-Ta | 3.05 | -1.32 | V-V | 2.96 | -0.55 | Nb-Nb | 3.04 | -1.10 | Nb-Nb | 3.07 | -1.00 |

| $Mo_2HfAlC_2$ | | | $Mo_2TaAlC_2$ | | | $Mo_2TiAlC_2$ | | |
|---|---|---|---|---|---|---|---|---|
| Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) |
| Mo-Mo | 3.07 | -0.81 | Mo-Mo | 3.06 | -0.84 | Mo-Mo | 2.99 | -0.96 |
| Mo-Hf | 3.14 | -0.88 | Mo-Ta | 3.05 | -1.02 | Mo-Ti | 3.04 | -0.67 |
| Mo-Al | 2.78 | -1.95 | Mo-Al | 2.81 | -1.84 | Mo-Al | 2.79 | -1.89 |
| Mo-C | 2.14 | -3.89 | Mo-C | 2.12 | -3.97 | Mo-C | 2.11 | -4.18 |
| Hf-C | 2.26 | -3.67 | Ta-C | 2.20 | -3.94 | Ti-C | 2.15 | -2.92 |
| Hf-Hf | 3.07 | -1.44 | Ta-Ta | 3.06 | -1.26 | Ti-Ti | 2.99 | -0.63 |

| $Mo_2VAlC_2$ | | | $Mo_2YAlC_2$ | | | $Mo_2ZrAlC_2$ | | | $Nb_2HfAlC_2$ | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) |
| Mo-Mo | 2.97 | -0.99 | Mo-Mo | 3.14 | -0.66 | Mo-Mo | 3.10 | -0.75 | Nb-Nb | 3.16 | -0.85 |
| Mo-V | 2.96 | -0.68 | Mo-Y | 3.29 | -0.70 | Mo-Zr | 3.16 | -0.77 | Nb-Hf | 3.17 | -0.97 |
| Mo-Al | 2.80 | -1.82 | Mo-Al | 2.77 | -2.05 | Mo-Al | 2.78 | -1.97 | Nb-Al | 2.87 | -1.88 |
| Mo-C | 2.09 | -4.27 | Mo-C | 2.12 | -4.16 | Mo-C | 2.13 | -4.01 | Nb-C | 2.18 | -3.97 |
| V-C | 2.10 | -3.01 | Y-C | 2.45 | -2.74 | Zr-C | 2.30 | -3.29 | Hf-C | 2.29 | -3.45 |
| V-V | 2.97 | -0.49 | Y-Y | 3.14 | -1.71 | Zr-Zr | 3.10 | -1.19 | Hf-Hf | 3.16 | -1.18 |

| $Nb_2ScAlC_2$ | | | $Nb_2TaAlC_2$ | | | $Nb_2TiAlC_2$ | | | $Nb_2VAlC_2$ | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) |
| Nb-Nb | 3.18 | -0.77 | Nb-Nb | 3.11 | -1.02 | Nb-Nb | 3.10 | -0.94 | Nb-Nb | 3.04 | -1.12 |
| Nb-Sc | 3.14 | -0.71 | Nb-Ta | 3.13 | -1.00 | Nb-Ti | 3.05 | -0.75 | Nb-V | 3.01 | -0.71 |
| Nb-Al | 2.90 | -1.80 | Nb-Al | 2.87 | -1.81 | Nb-Al | 2.87 | -1.84 | Nb-Al | 2.90 | -1.73 |
| Nb-C | 2.14 | -4.49 | Nb-C | 2.19 | -3.82 | Nb-C | 2.16 | -4.30 | Nb-C | 2.15 | -4.21 |
| Sc-C | 2.33 | -2.27 | Ta-C | 2.21 | -3.87 | Ti-C | 2.19 | -2.68 | V-C | 2.12 | -2.86 |
| Sc-Sc | 3.18 | -0.57 | Ta-Ta | 3.11 | -1.13 | Ti-Ti | 3.10 | -0.49 | V-V | 3.04 | -0.42 |

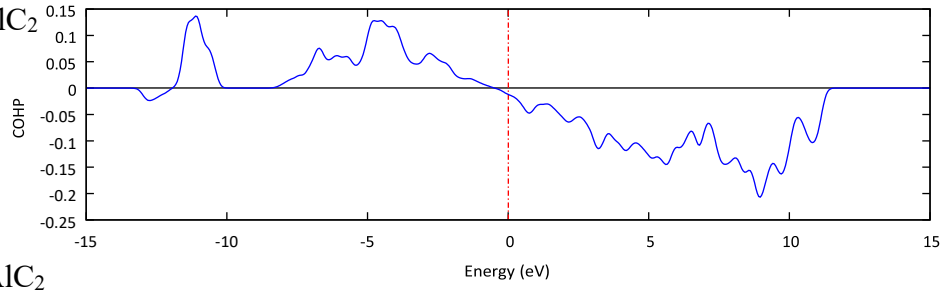
| $Nb_2YAlC_2$ | | | $Nb_2ZrAlC_2$ | | | $W_2HfAlC_2$ | | |
|---|---|---|---|---|---|---|---|---|
| Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) |
| Nb-Nb | 3.25 | -0.65 | Nb-Nb | 3.18 | -0.80 | W-W | 3.05 | -1.10 |
| Nb-Y | 3.30 | -0.79 | Nb-Zr | 3.90 | -0.83 | W-Hf | 3.16 | -0.98 |
| Nb-Al | 2.90 | -1.83 | Nb-Al | 2.87 | -1.88 | W-Al | 2.80 | -2.25 |
| Nb-C | 2.16 | -4.28 | Nb-C | 2.18 | -4.09 | W-C | 2.15 | -4.12 |
| Y-C | 2.49 | -2.53 | Zr-C | 2.34 | -3.08 | Hf-C | 2.25 | -3.69 |
| Y-Y | 3.25 | -1.36 | Zr-Zr | 3.18 | -1.01 | Hf-Hf | 3.05 | -1.55 |

| $W_2TiAlC_2$ | | | $W_2YAlC_2$ | | | $W_2ZrAlC_2$ | | |
|---|---|---|---|---|---|---|---|---|
| Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) | Bonding interaction | Bond length(A) | ICOHP (eV) |
| W-W | 2.98 | -1.26 | W-W | 3.12 | -0.91 | W-W | 3.08 | -0.98 |
| W-Ti | 3.06 | -0.86 | W-Y | 3.35 | -0.77 | W-Zr | 3.17 | -0.94 |
| W-Al | 2.79 | -2.25 | W-Al | 2.79 | -2.35 | W-Al | 2.78 | -2.36 |
| W-C | 2.12 | -4.41 | W-C | 2.15 | -4.27 | W-C | -2.13 | -4.26 |
| Ti-C | 2.15 | -2.94 | Y-C | 2.45 | -2.76 | Zr-C | 2.29 | -3.33 |
| Ti-Ti | 2.98 | -0.68 | Y-Y | 3.12 | -1.78 | Zr-Zr | 3.08 | -1.26 |

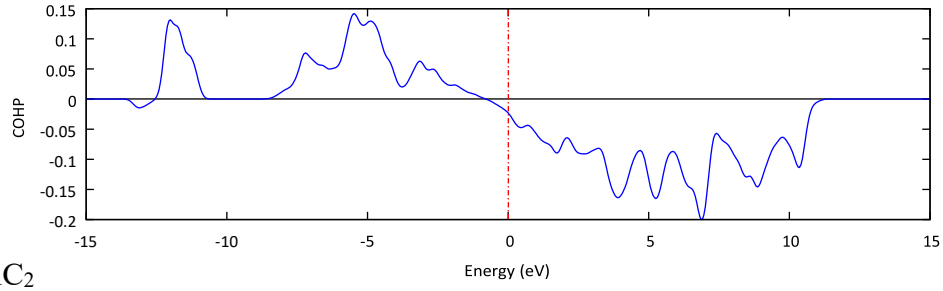
**Table 3S.** Superconductivity transition temperature for $\mu^*$ value 0.1 and 0.13, logarithmic average frequency and electron–phonon coupling parameters ($\boldsymbol{\lambda}$).

| **Material** | $\mathbf{T_c(K)}$, $(\boldsymbol{\mu^* = 0.1 - 13})$ | $\boldsymbol{\omega_{log}(K)}$ | $\boldsymbol{\lambda}$ |
|---|---|---|---|
| $Mo_2HfAlC_2$ | 2.5-1.37 | 274.5 | 0.46 |
| $Mo_2NbAlC_2$ | 11.0-8.7 | 273.1 | 0.74 |
| $Mo_2ScAlC_2$ | 3.6-2.1 | 327.2 | 0.48 |
| $Mo_2TaAlC_2$ | 8.8-6.7 | 258.4 | 0.69 |
| $Mo_2TiAlC_2$ | 2.7-1.4 | 314.9 | 0.46 |
| $Mo_2VAlC_2$ | 8.0-5.7 | 316.1 | 0.62 |
| $Mo_2YAlC_2$ | 6.8-4.8 | 280.3 | 0.61 |
| $Mo_2ZrAlC_2$ | 3.9-2.4 | 280.2 | 0.51 |
| **Material** | $\mathbf{T_c(K)}$, $(\boldsymbol{\mu^* = 0.1 - 13})$ | $\boldsymbol{\omega_{log}(K)}$ | $\boldsymbol{\lambda}$ |
| $Nb_2HfAlC_2$ | 0.5-0.1 | 365.1 | 0.34 |
| $Nb_2ScAlC_2$ | 0.0-0.0 | 413.8 | 0.26 |
| $Nb_2TaAlC_2$ | 0.0-0.0 | 350.9 | 0.21 |
| $Nb_2TiAlC_2$ | 0.5-0.1 | 371.3 | 0.34 |
| $Nb_2VAlC_2$ | 0.9-0.3 | 388.5 | 0.36 |
| $Nb_2YAlC_2$ | 0.3-0.07 | 381.4 | 0.32 |
| $Nb_2ZrAlC_2$ | 0.3-0.07 | 379.8 | 0.31 |
| **Material** | $\mathbf{T_c(K)}$, $(\boldsymbol{\mu^* = 0.1 - 13})$ | $\boldsymbol{\omega_{log}(K)}$ | $\boldsymbol{\lambda}$ |
| $W_2HfAlC_2$ | 3.3-2.1 | 231.1 | 0.52 |
| $W_2NbAlC_2$ | 14.8-12.5 | 215.4 | 0.99 |
| $W_2ScAlC_2$ | 0.1-0.03 | 296.5 | 0.3 |
| $W_2TaAlC_2$ | 14.5-12.45 | 198.3 | 1.02 |
| $W_2TiAlC_2$ | 1.8-0.9 | 269.2 | 0.43 |
| $W_2VAlC_2$ | 17.9-15.1 | 259.6 | 0.98 |
| $W_2YAlC_2$ | 1.9-1.1 | 261.5 | 0.44 |
| $W_2ZrAlC_2$ | 4.3-2.8 | 231.7 | 0.56 |

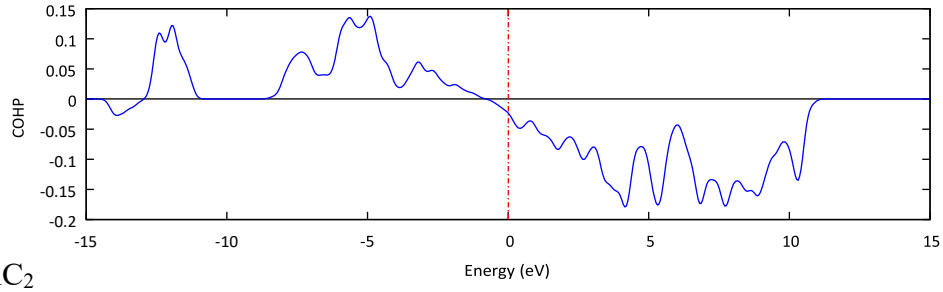
**a)** $Mo_2HfAlC_2$



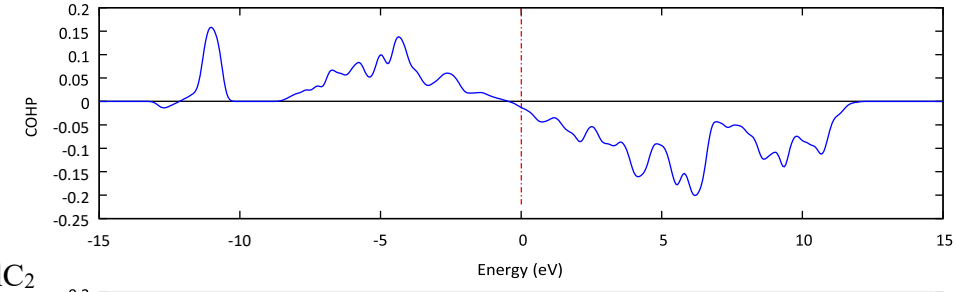

**b)** $Mo_2NbAlC_2$



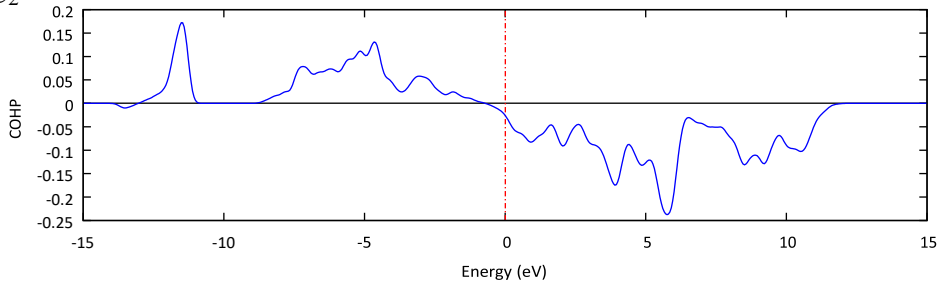

**c)** $Mo_2TaAlC_2$



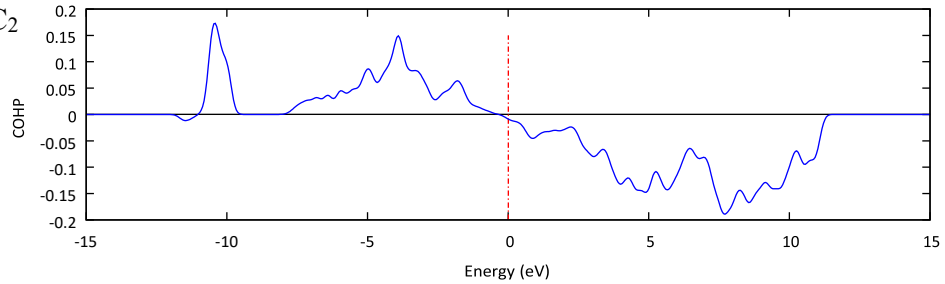

**d)** $Mo_2TiAlC_2$



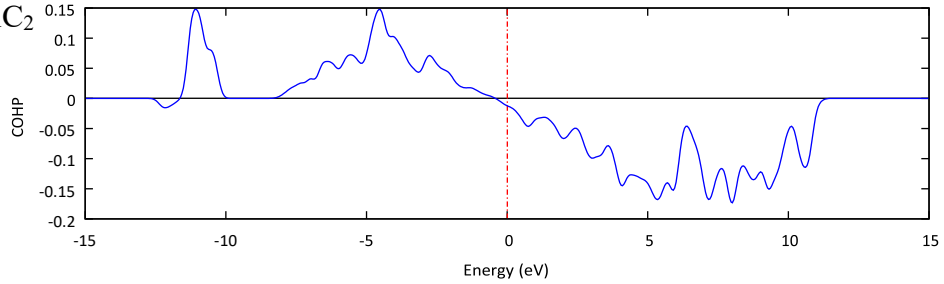

**e)** $Mo_2VAlC_2$



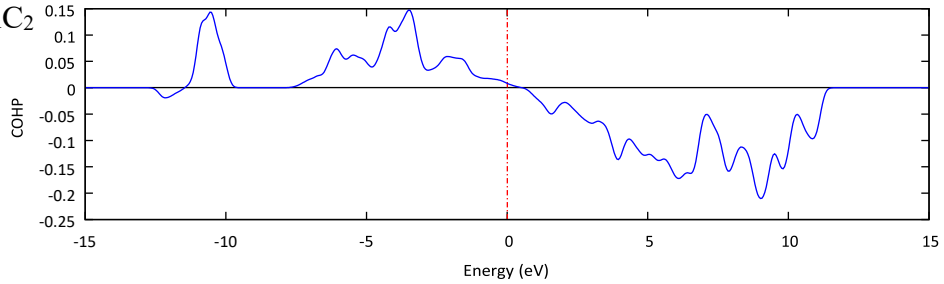
**f)** $Mo_2YAlC_2$



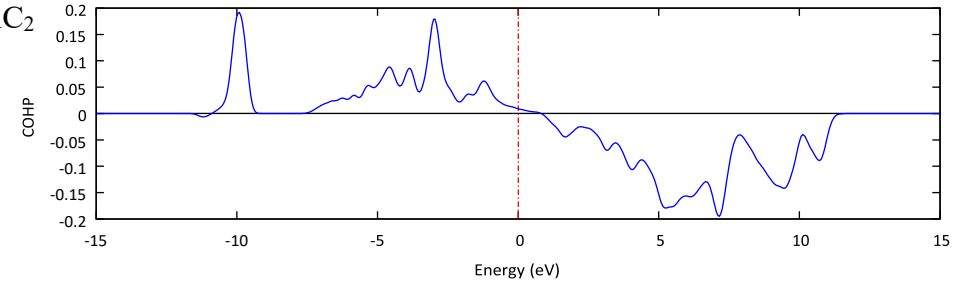

**g)** $Mo_2ZrAlC_2$



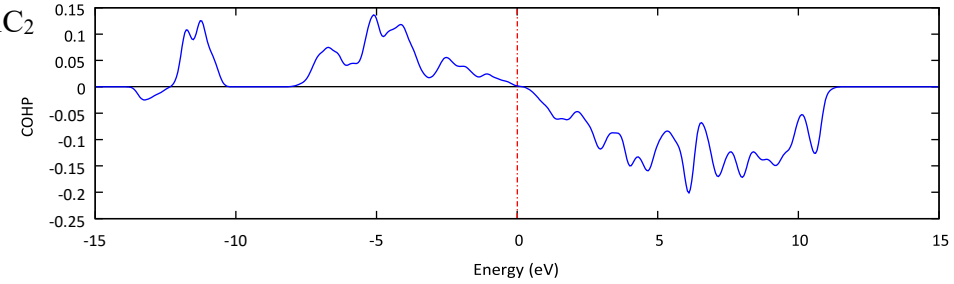

**h)** $Nb_2HfAlC_2$



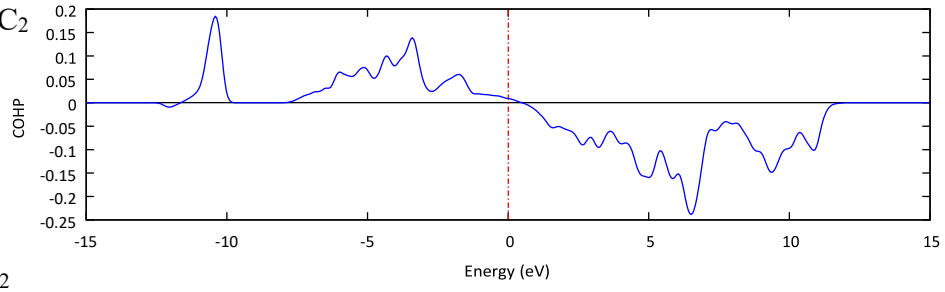

**i)** $Nb_2ScAlC_2$



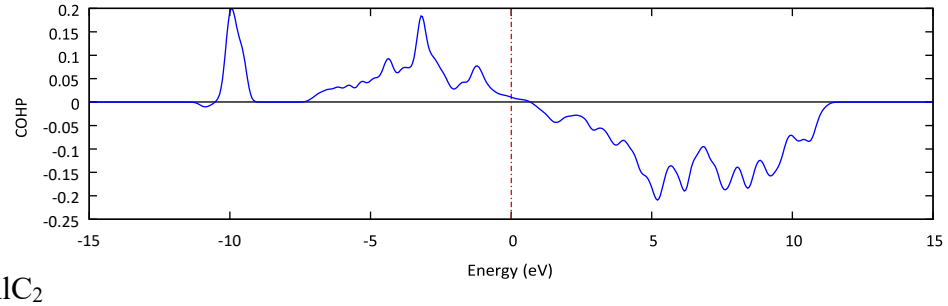

**j)** $Nb_2TaAlC_2$



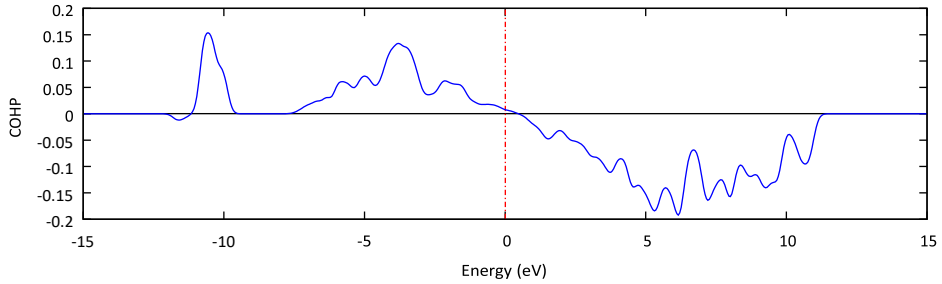
**k)** $Nb_2TiAlC_2$



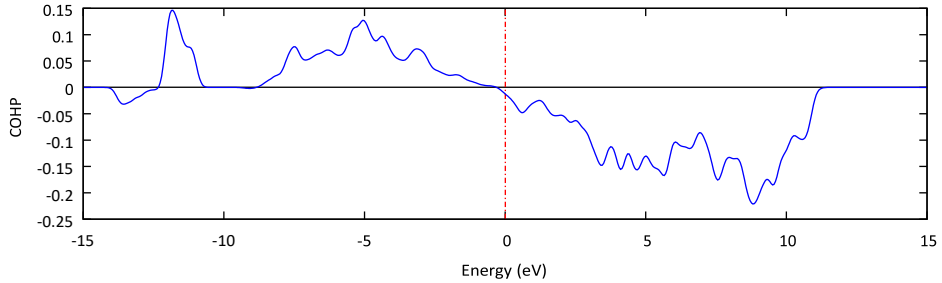

**l)** $Nb_2YAlC_2$



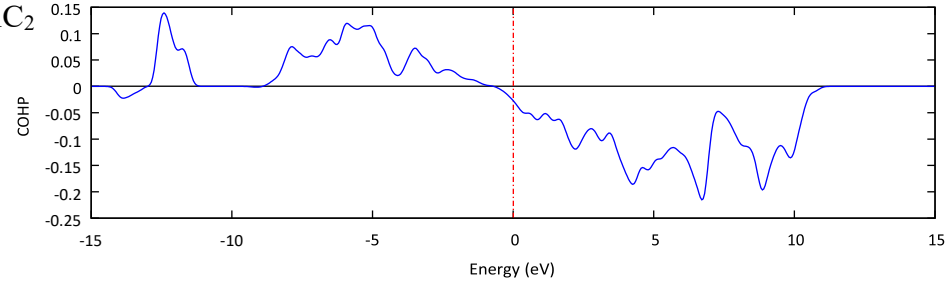

**m)** $Nb_2ZrAlC_2$




**n)** $W_2HfAlC_2$




**o)** $W_2NbAlC_2$



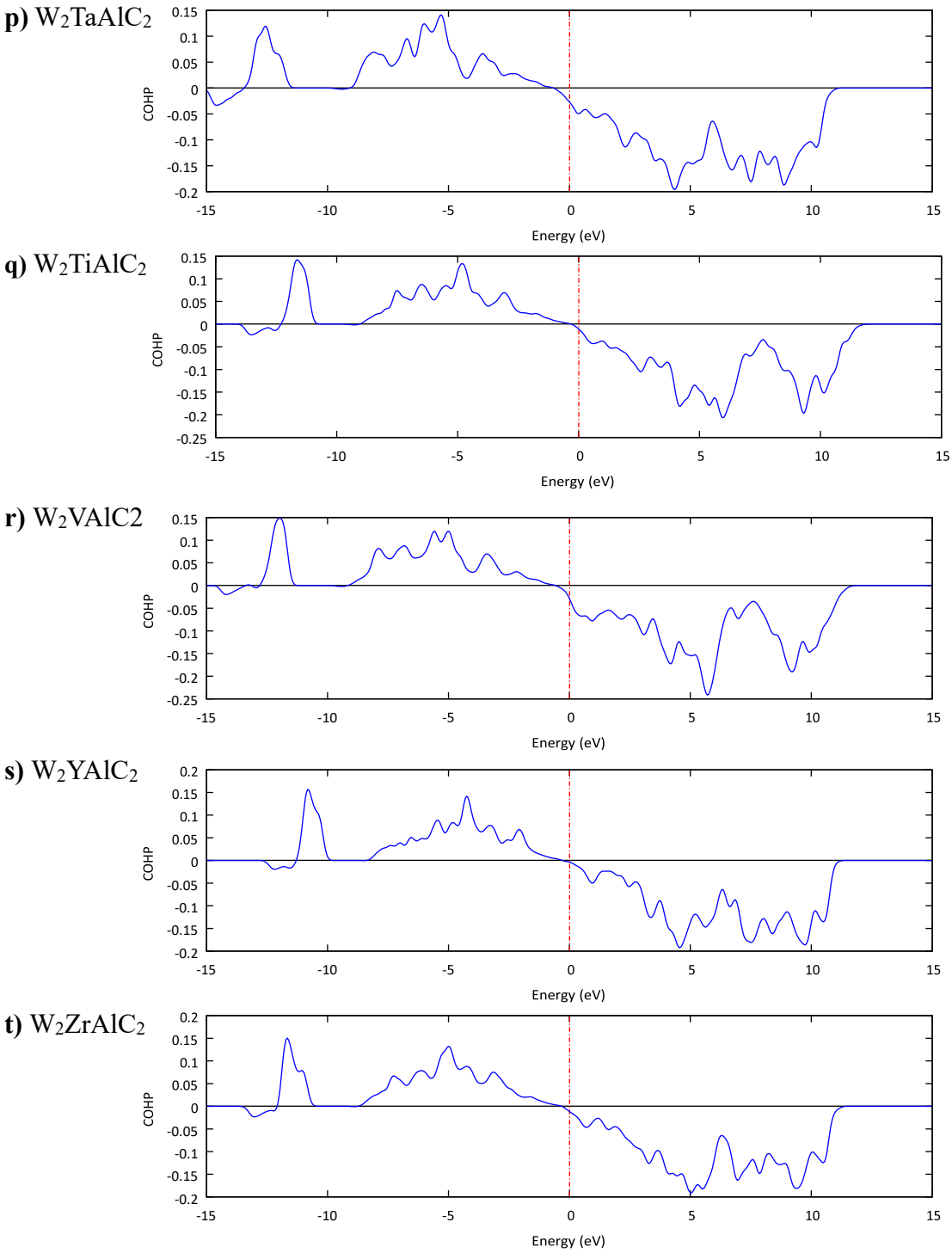
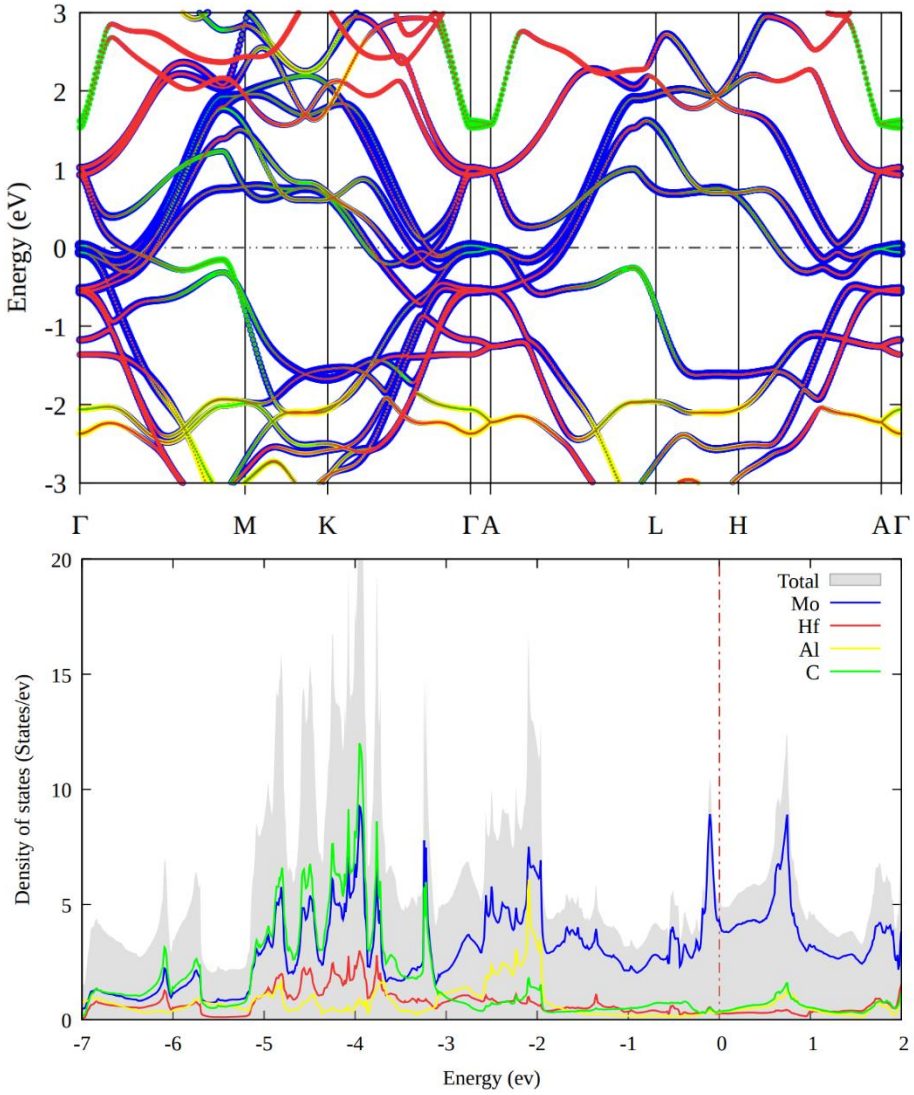
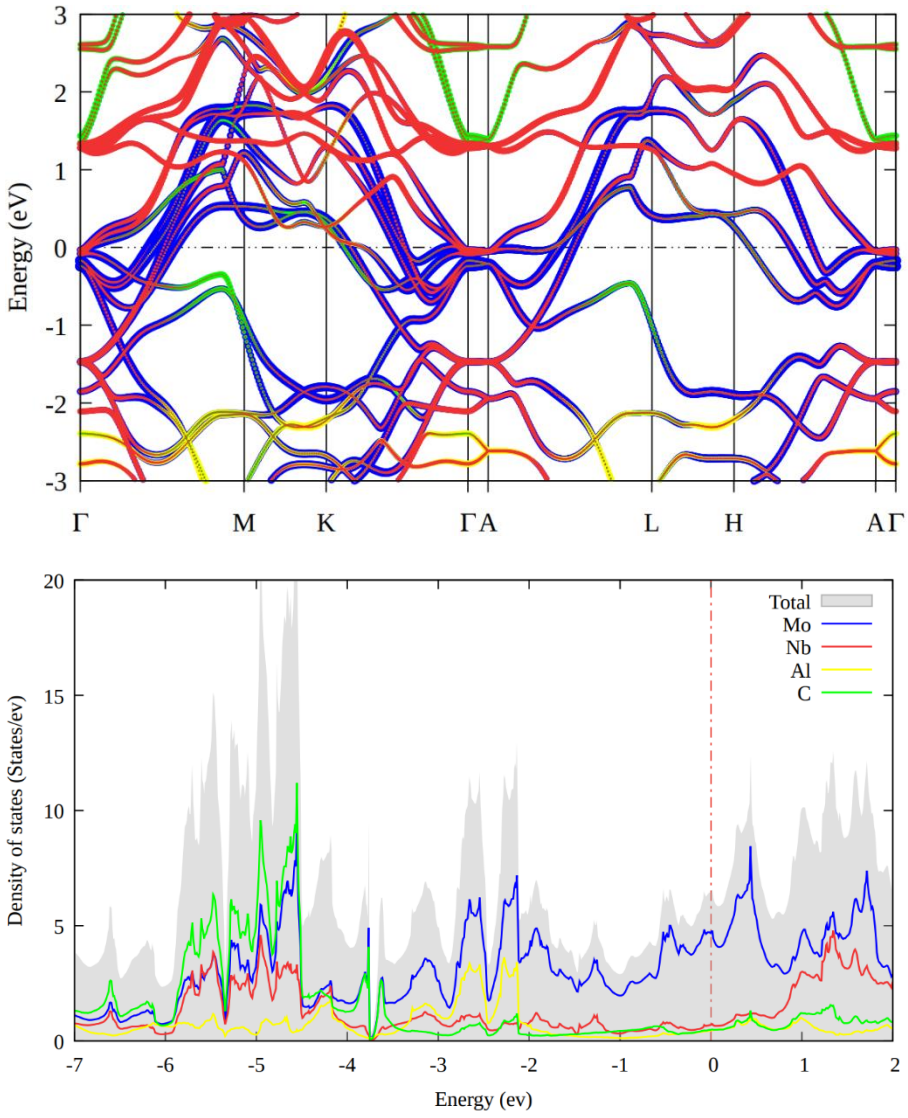



**Fig1S**. Crystal Orbital Hamilton Population (COHP) analysis of **a)** $Mo_2HfAlC_2$ **b)** $Mo_2NbAlC_2$ **c)** $Mo_2TaAlC_2$ **d)** $Mo_2TiAlC_2$ **e)** $Mo_2VAlC_2$ **f)** $Mo_2YAlC_2$ **g)** $Mo_2ZrAlC_2$ **h)** $Nb_2HfAlC_2$ **i)** $Nb_2ScAlC_2$ **j)** $Nb_2TaAlC_2$ **k)** $Nb_2TiAlC_2$ **l)** $Nb_2YAlC_2$ **m)** $Nb_2ZrAlC_2$ **n)** $W_2HfAlC_2$ **o)** $W_2NbAlC_2$ **p)** $W_2TaAlC_2$ **q)** $W_2TiAlC_2$ **r)** $W_2VAlC2$ **s)** $W_2YAlC_2$ **t)** $W_2ZrAlC_2$.

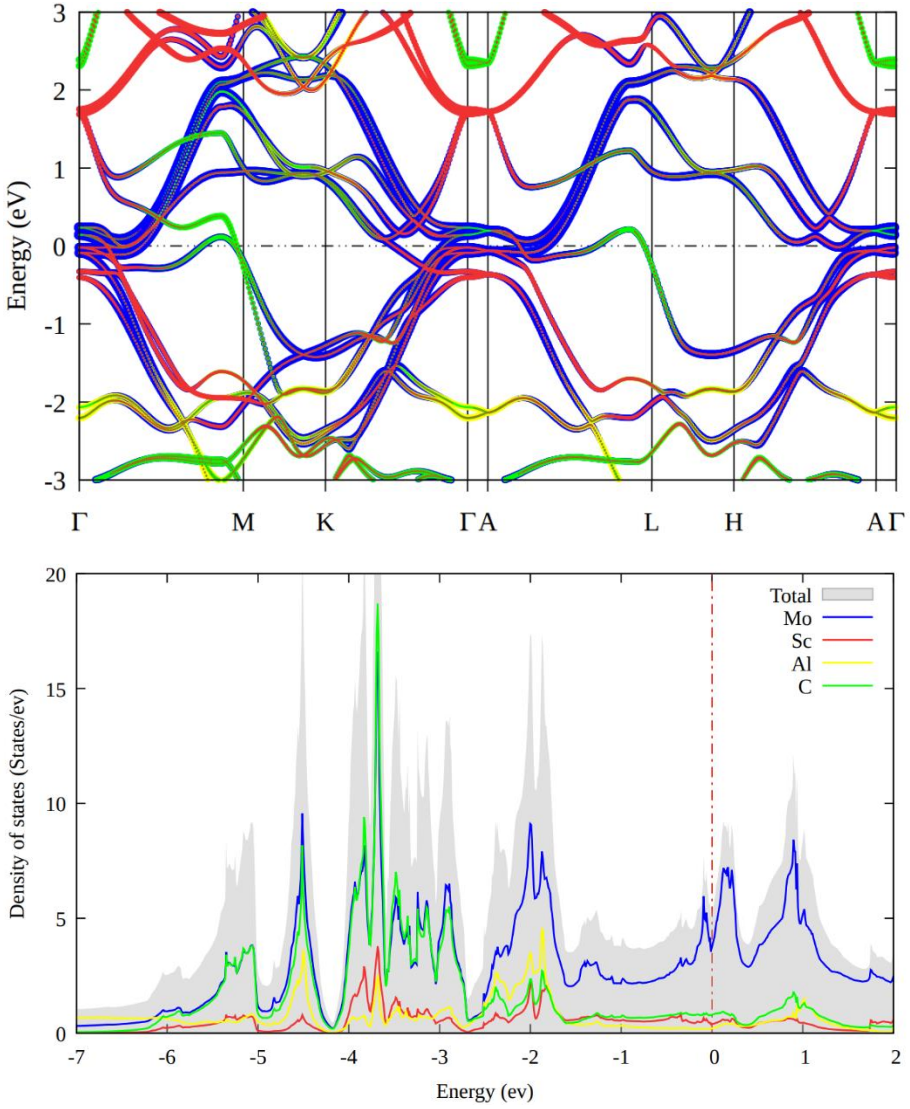
**a)** $Mo_2HfAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Mo
Hf
Al
C

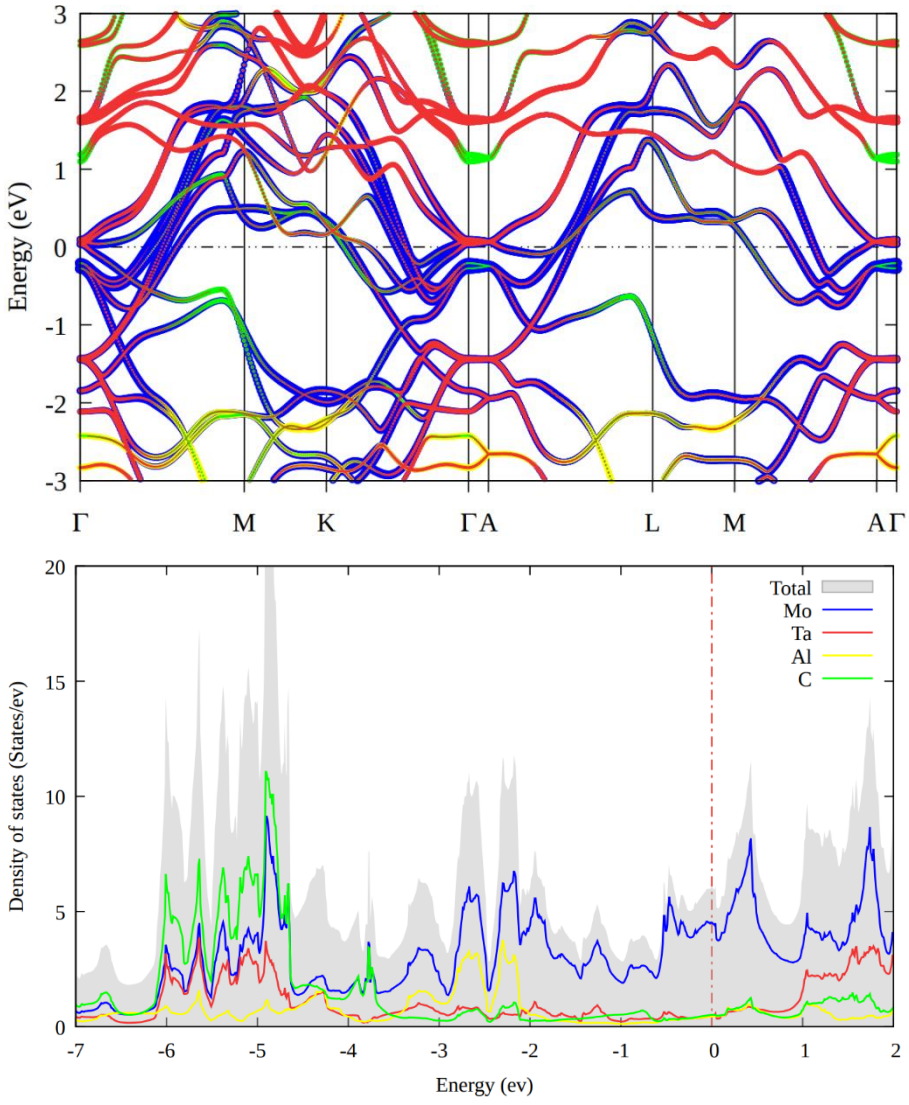
**b)** $Mo_2NbAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Mo
Nb
Al
C

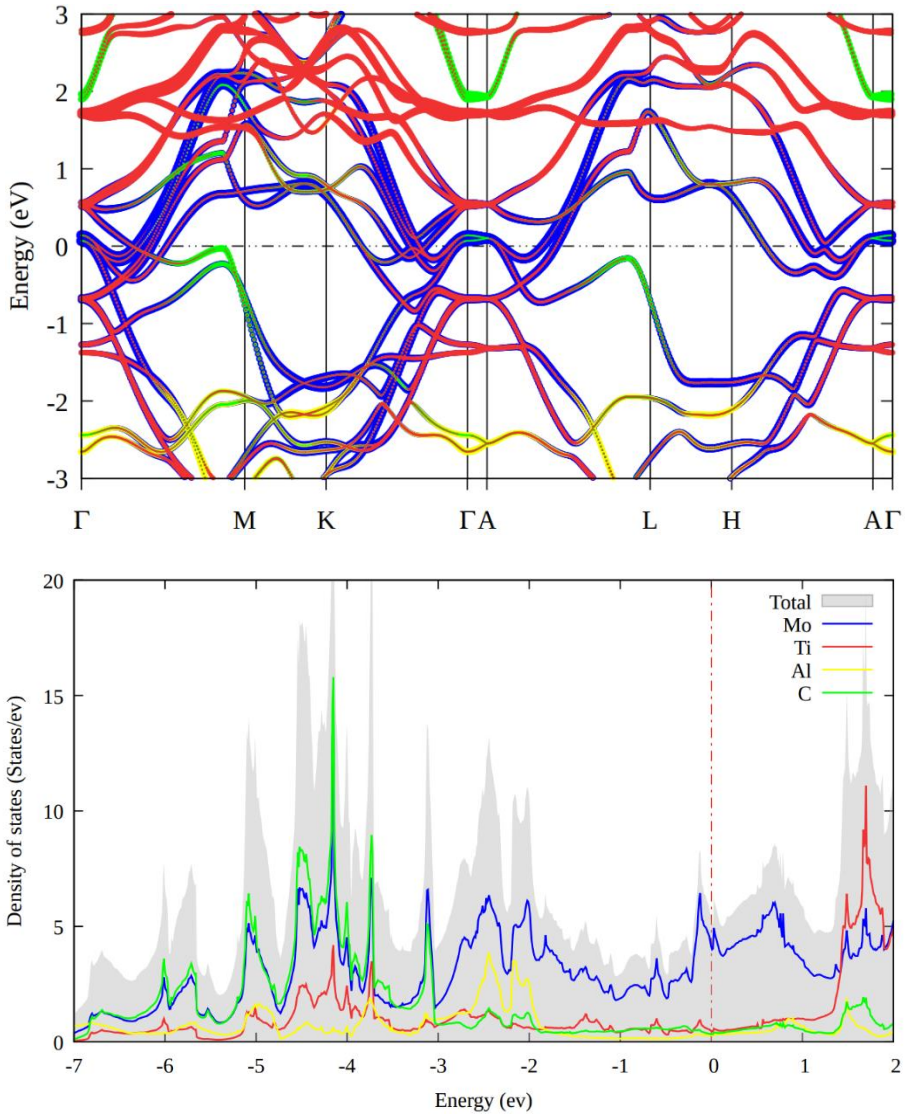
**c)** $Mo_2ScAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Mo
Sc
Al
C

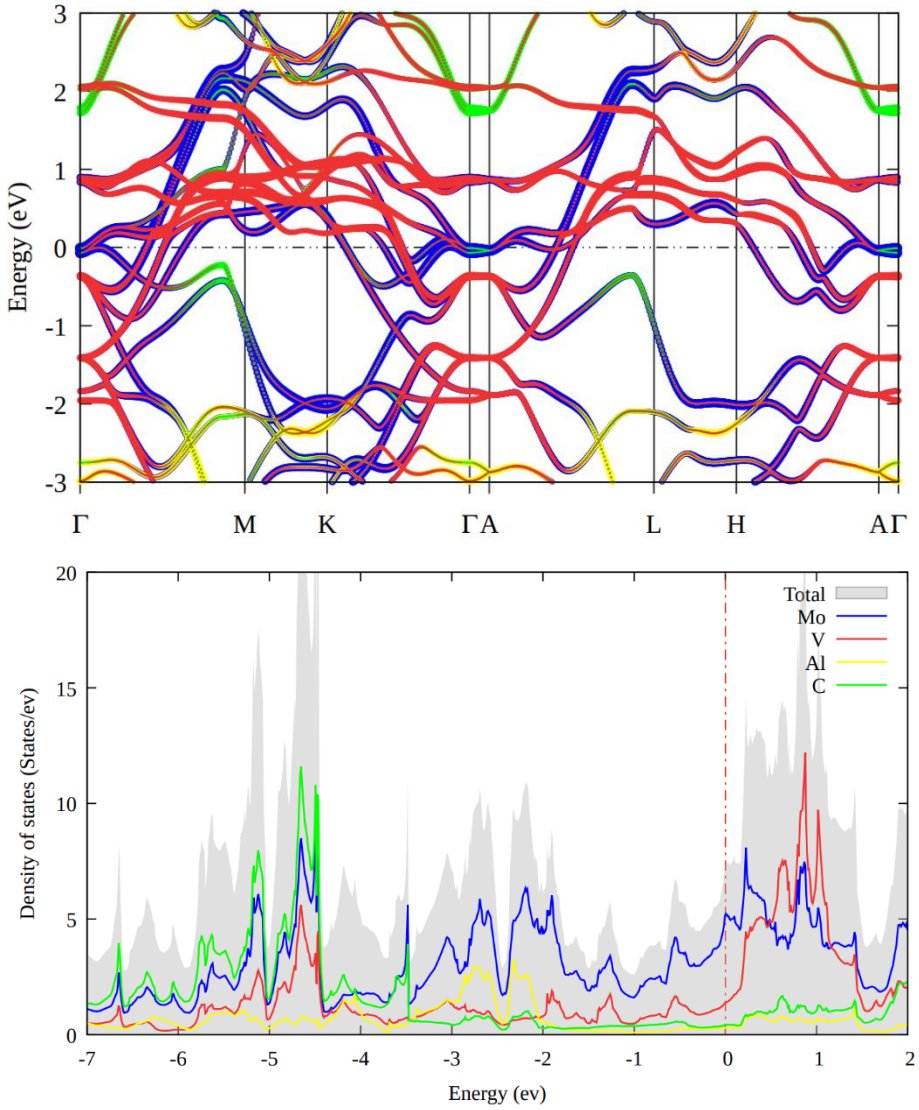
**d)** $Mo_2TaAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
M
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Mo
Ta
Al
C

**e)** $Mo_2TiAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Mo
Ti
Al
C

**f)** $Mo_2VAlC_2$


Energy (eV)
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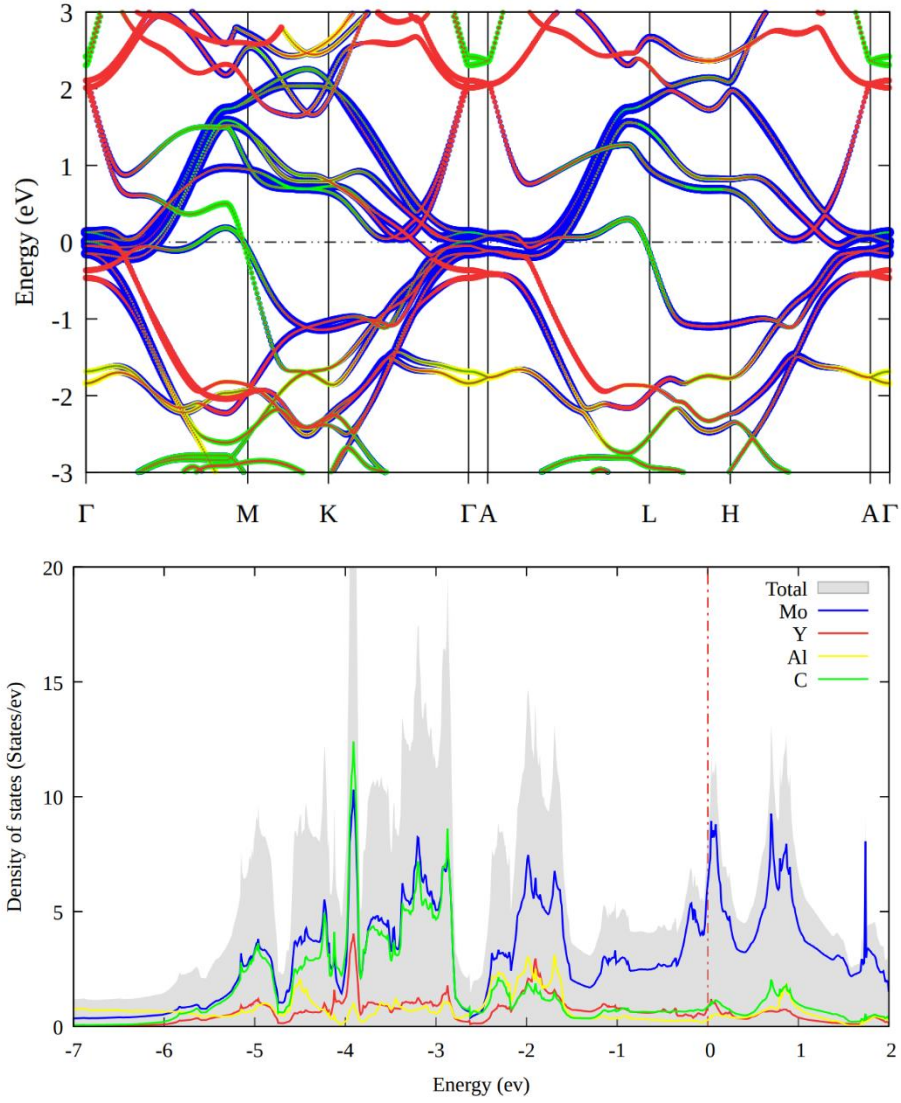
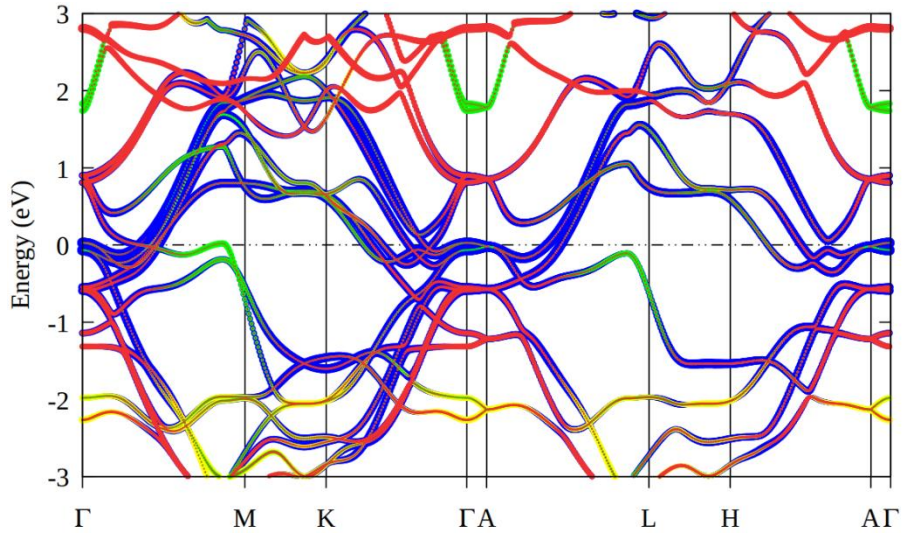
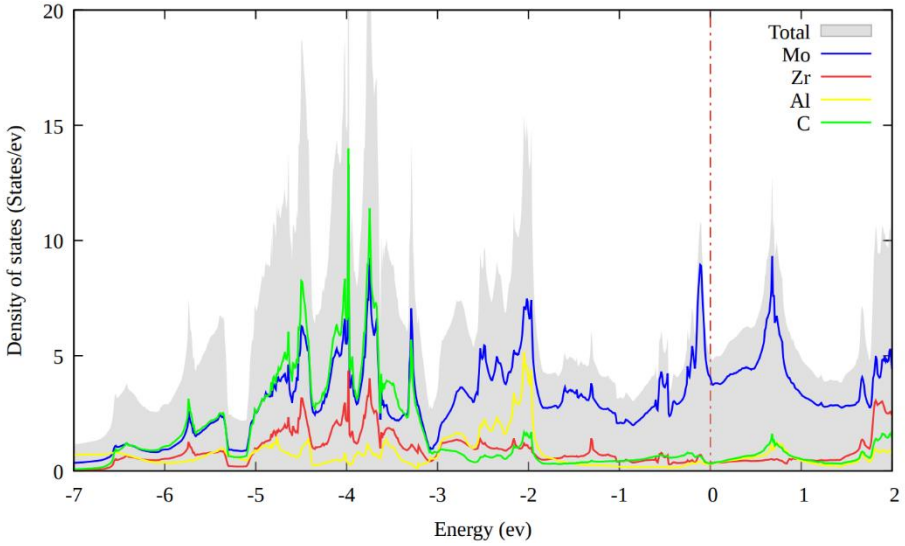
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Mo
V
Al
C

**g)** $Mo_2YAlC_2$

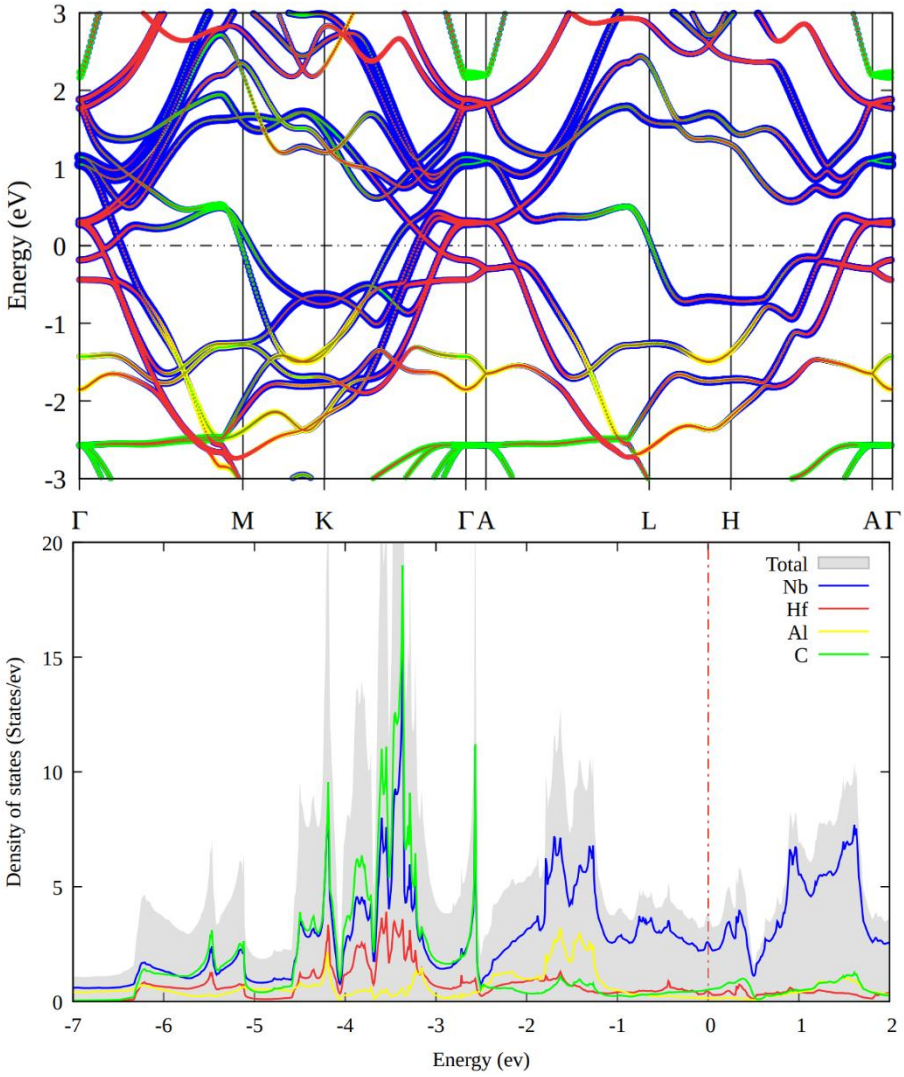

Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Mo
Y
Al
C

**h)** $Mo_2ZrAlC_2$

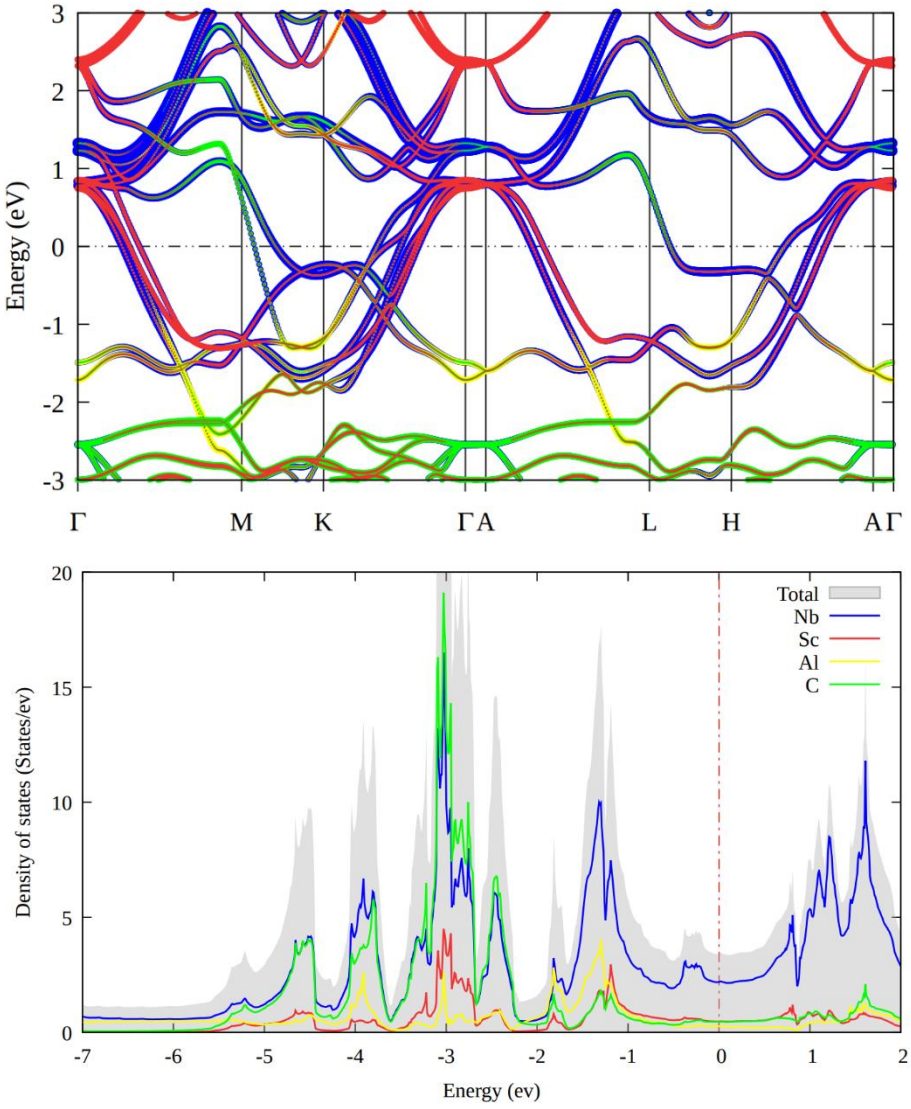

Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ


Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Mo
Zr
Al
C

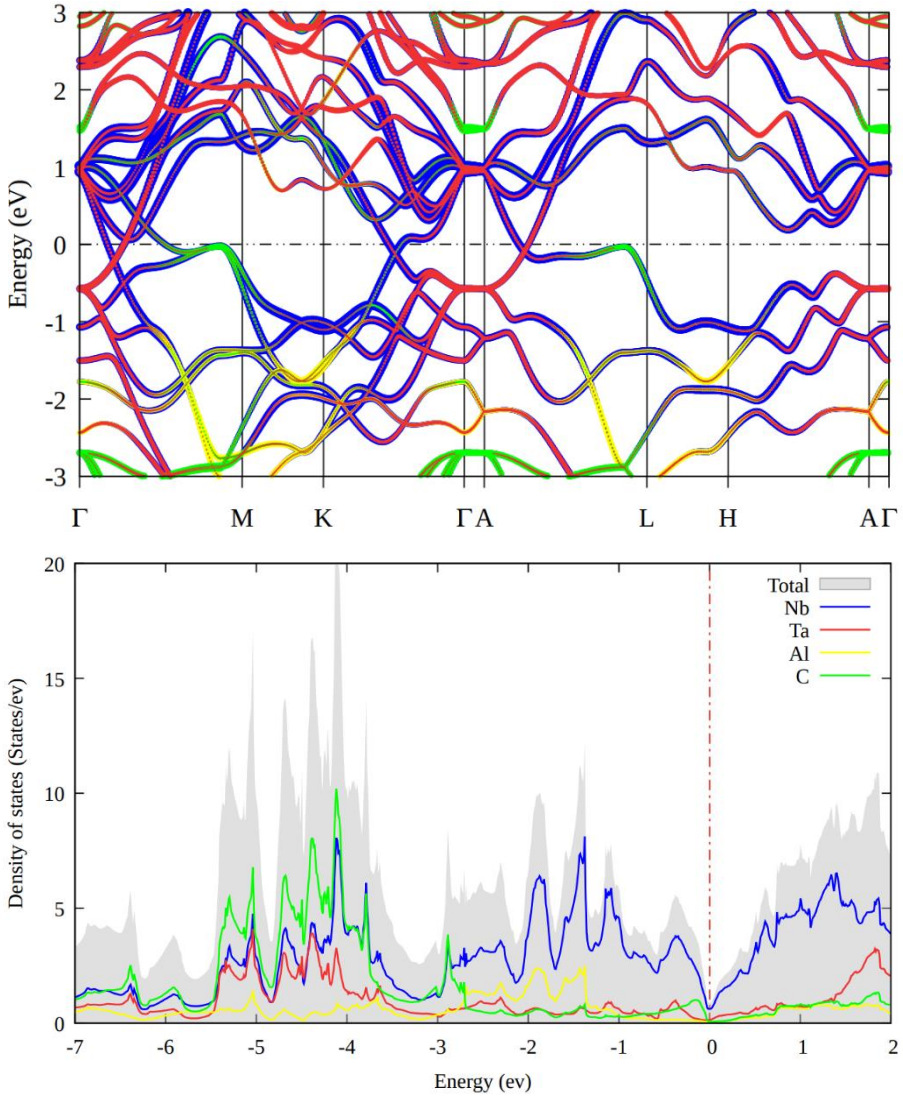
**i)** $Nb_2HfAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Nb
Hf
Al
C

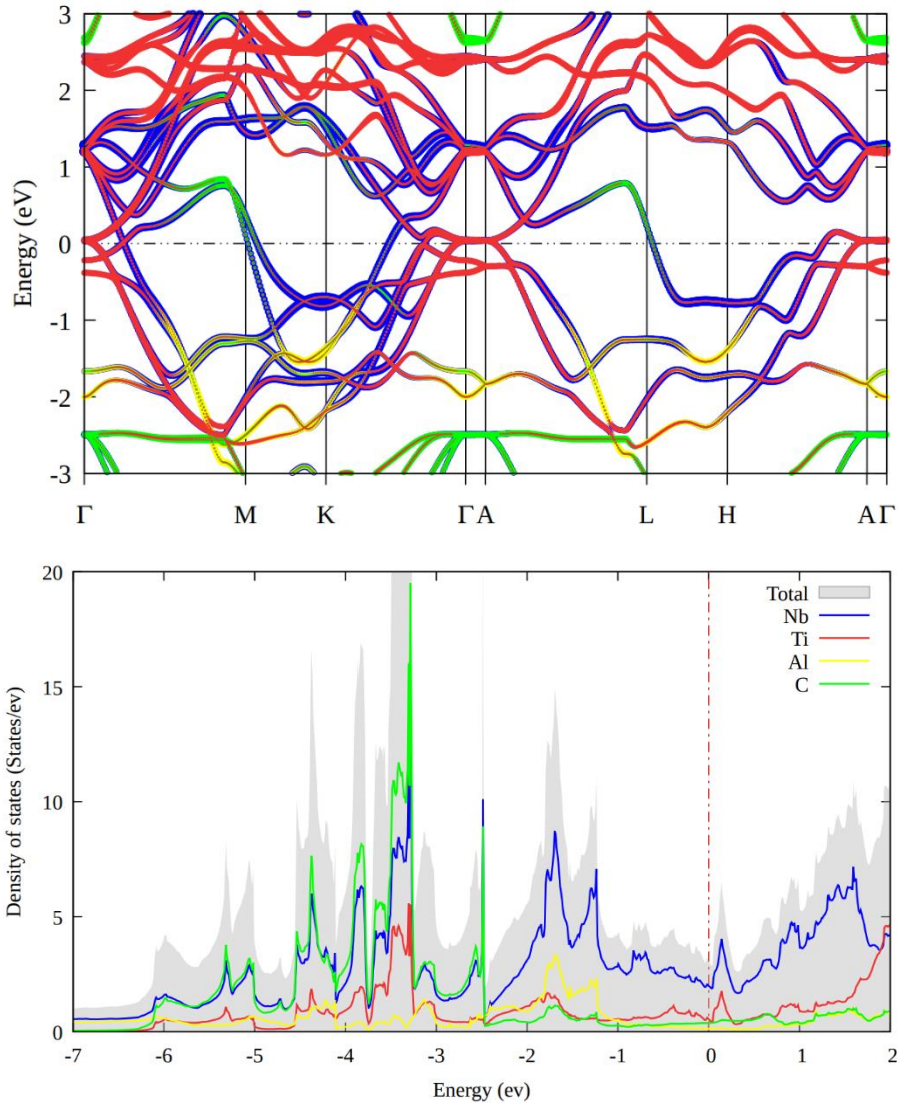
**j)** $Nb_2ScAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Nb
Sc
Al
C

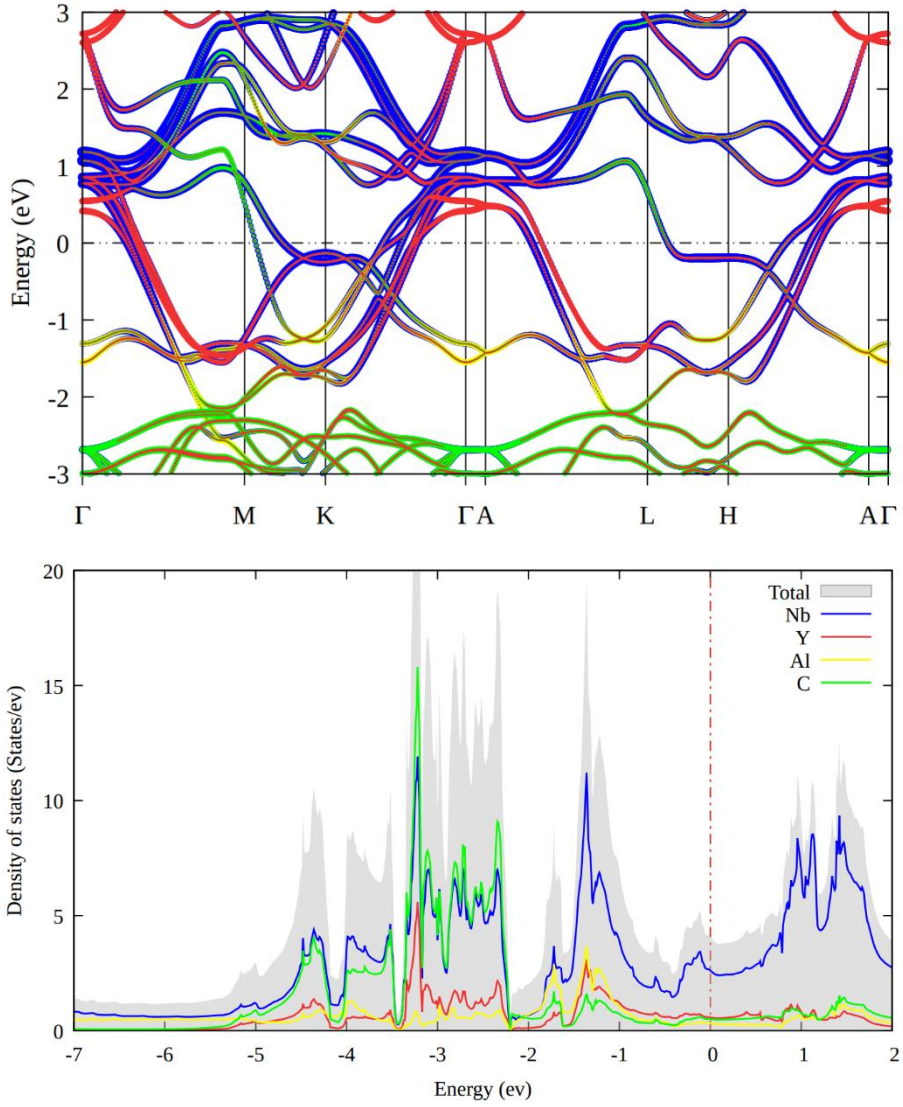
**k)** $Nb_2TaAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Nb
Ta
Al
C

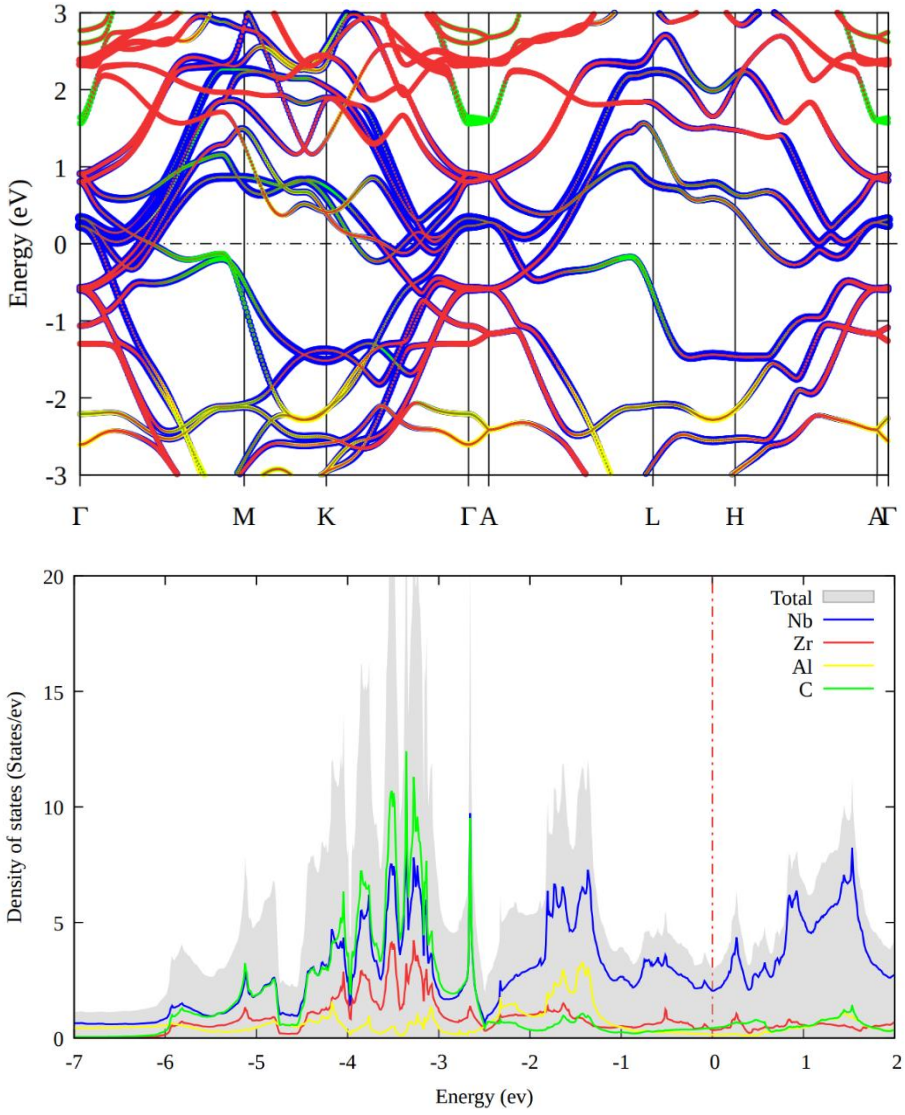
**I)** $Nb_2TiAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Nb
Ti
Al
C

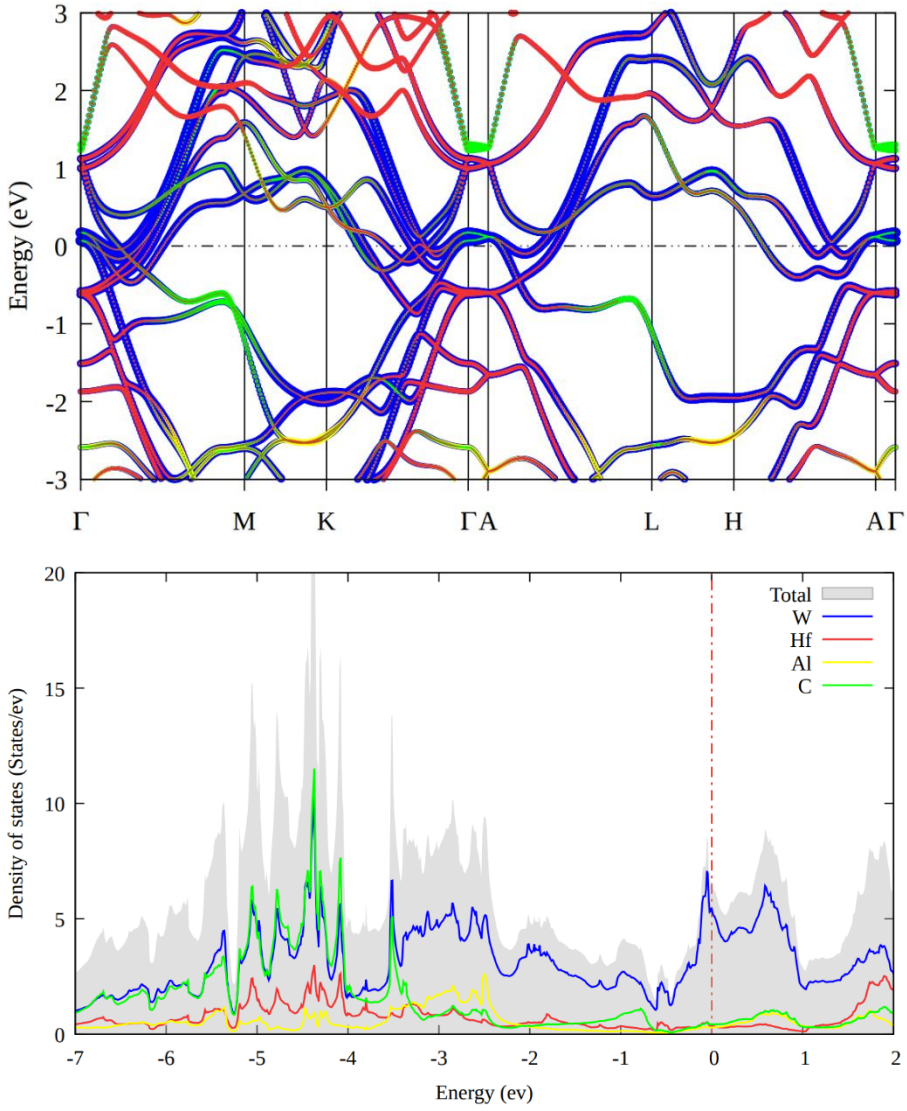
**m)** $Nb_2YAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Nb
Y
Al
C

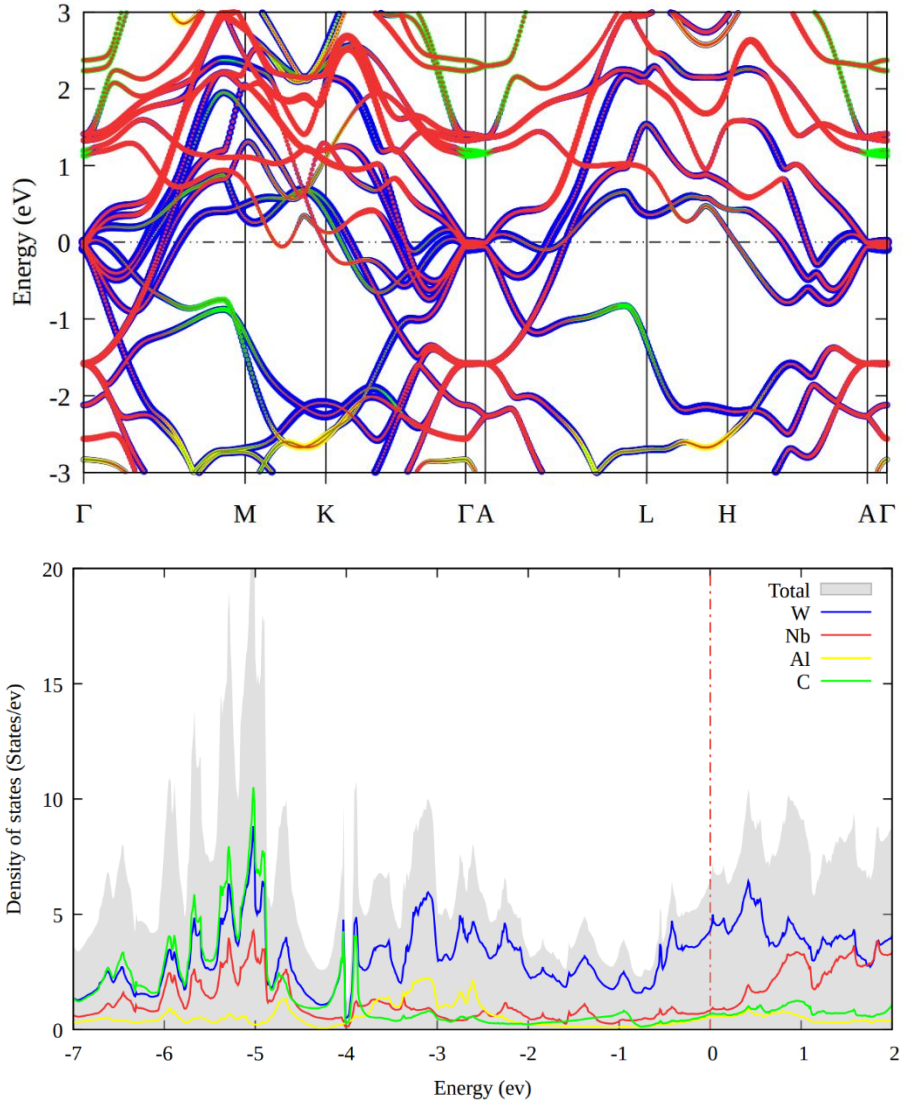
**n)** $Nb_2ZrAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
Nb
Zr
Al
C

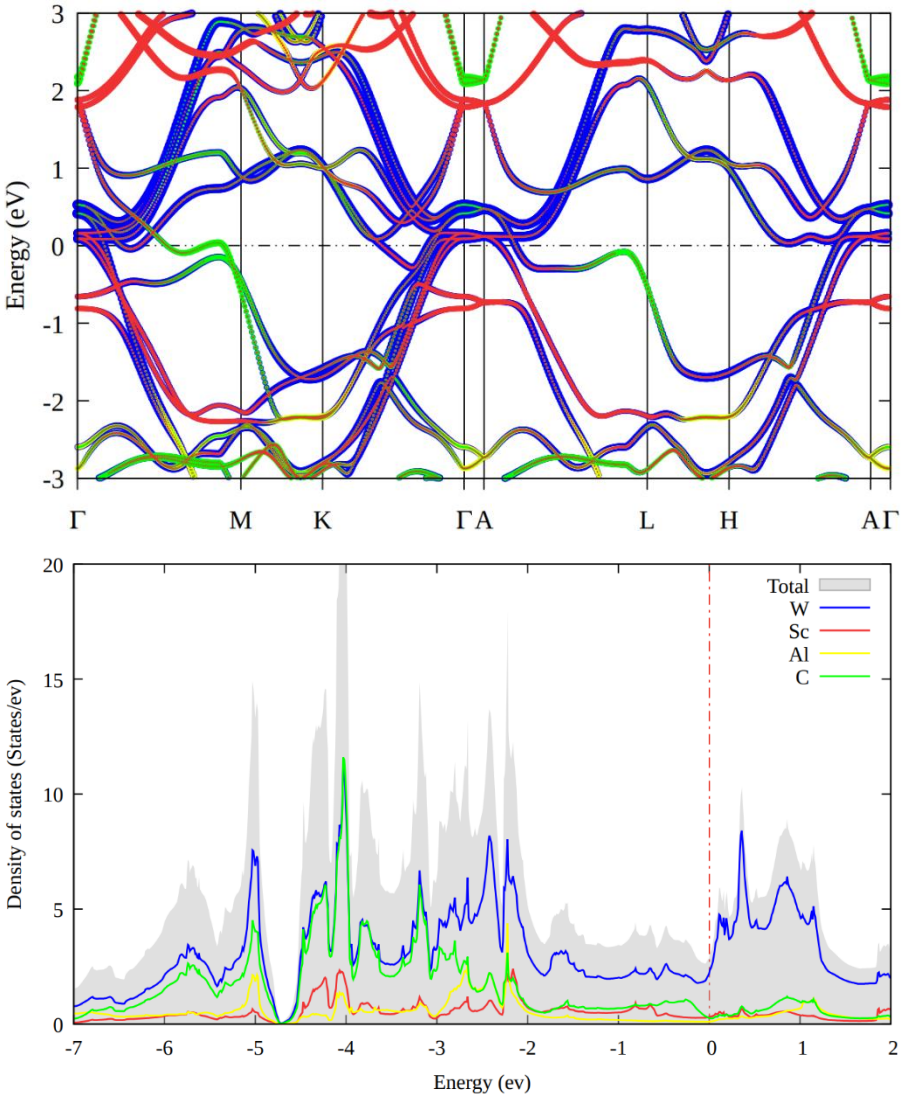
**o)** $W_2HfAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
W
Hf
Al
C

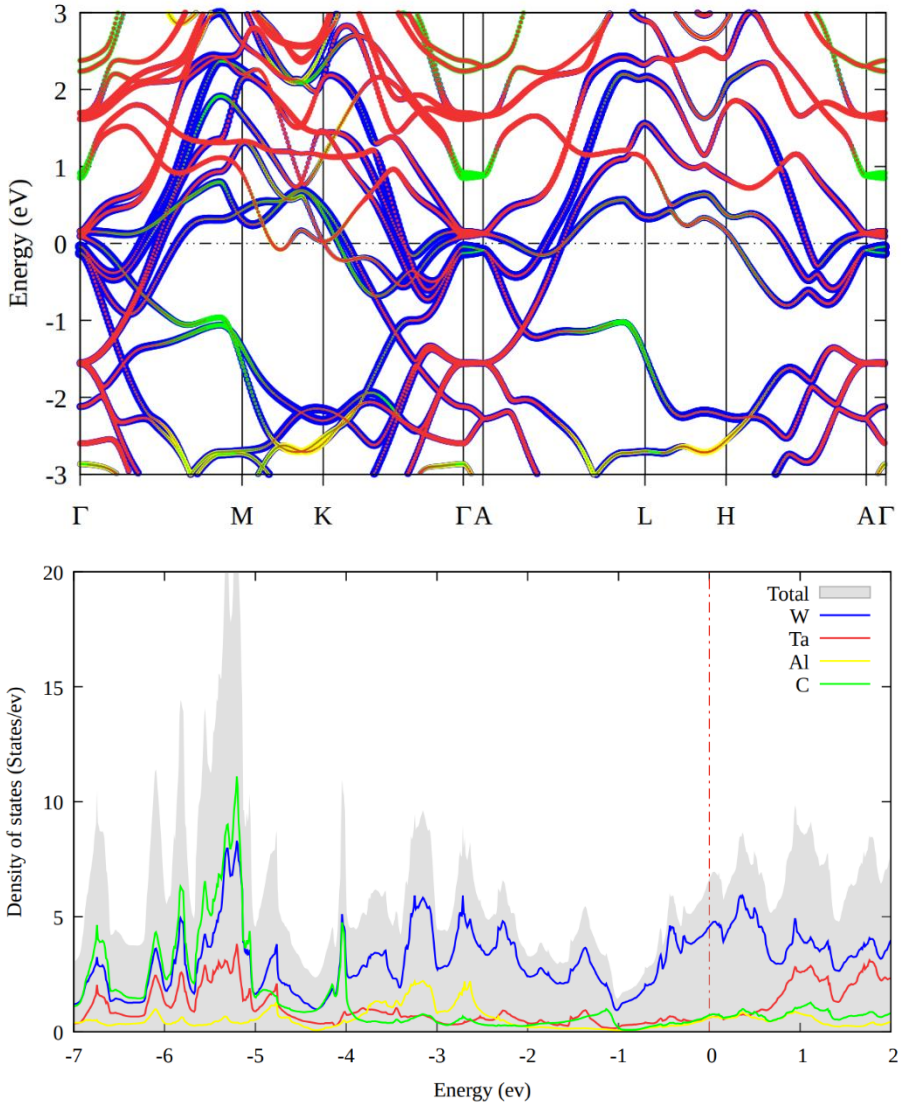
**p)** $W_2NbAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
W
Nb
Al
C

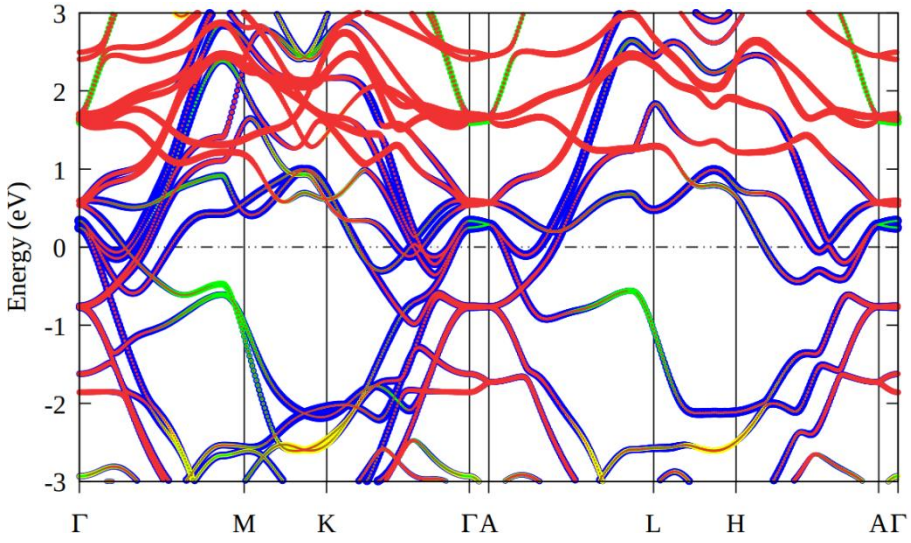
**q)** $W_2ScAlC_2$

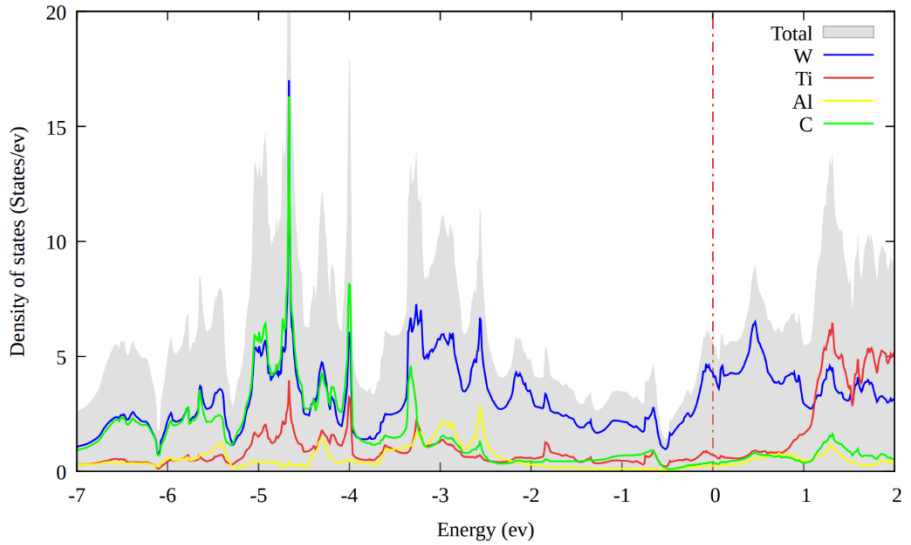

Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
W
Sc
Al
C

**r)** $W_2TaAlC_2$

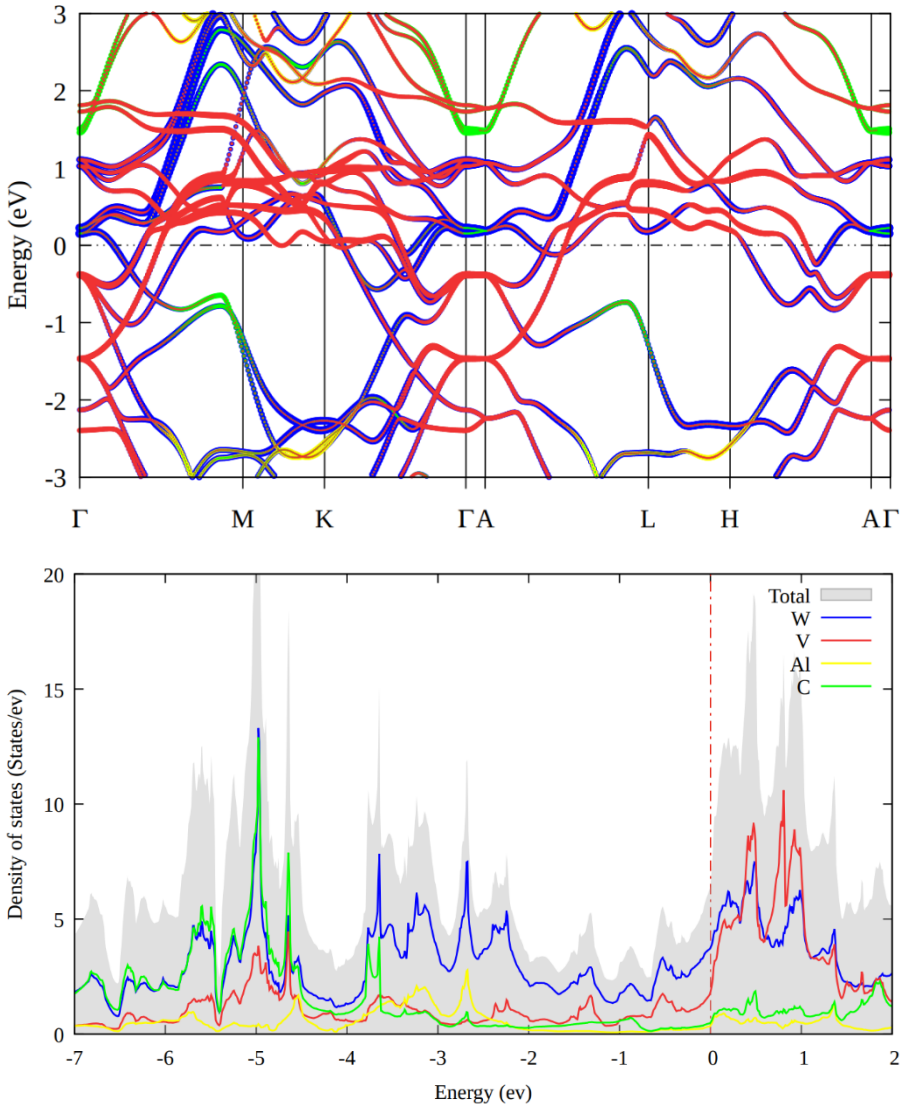

Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
W
Ta
Al
C

**s)** $W_2TiAlC_2$

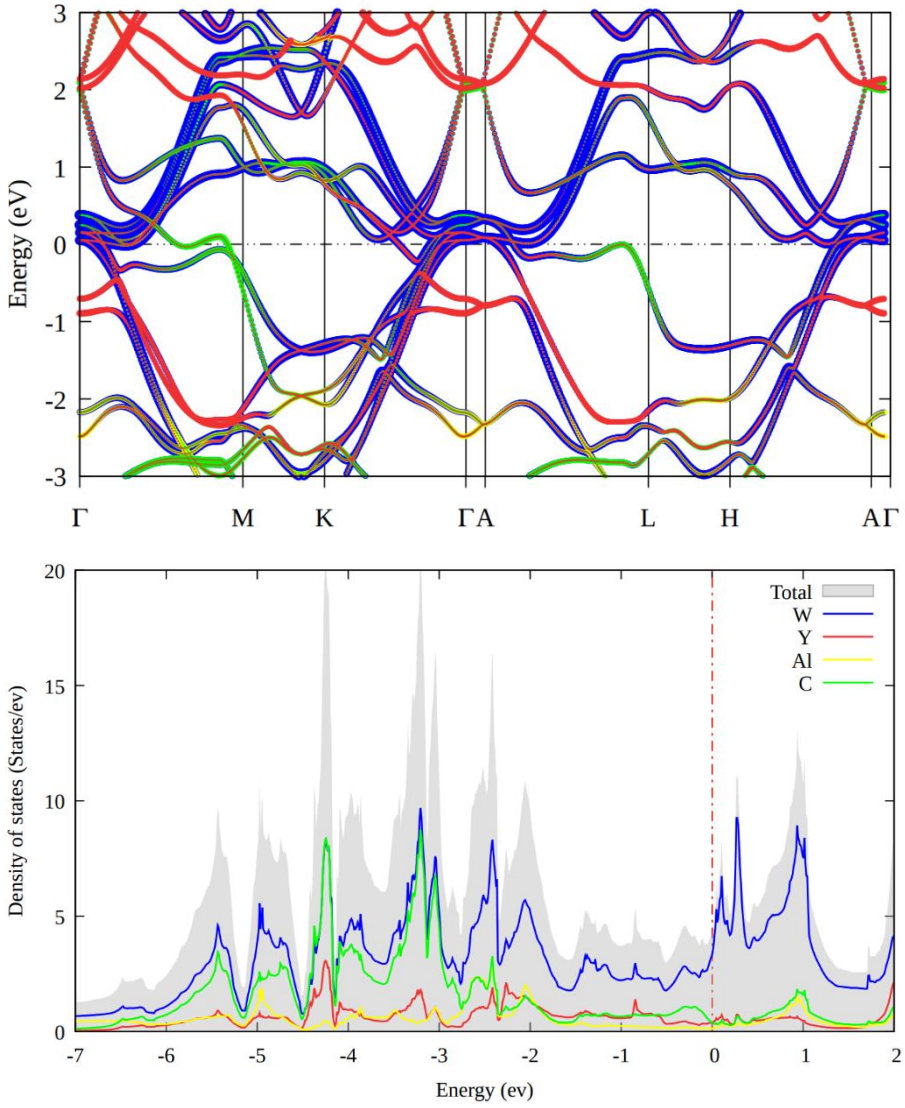

Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ


Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
W
Ti
Al
C

**t)** $W_2VAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
W
V
Al
C

**u)** $W_2YAlC_2$


Energy (eV)
3
2
1
0
-1
-2
-3
Γ
M
K
ΓA
L
H
AΓ
Density of states (States/ev)
20
15
10
5
0
-7
-6
-5
-4
-3
-2
-1
0
1
2
Energy (ev)
Total
W
Y
Al
C

**v)** $W_2ZrAlC_2$

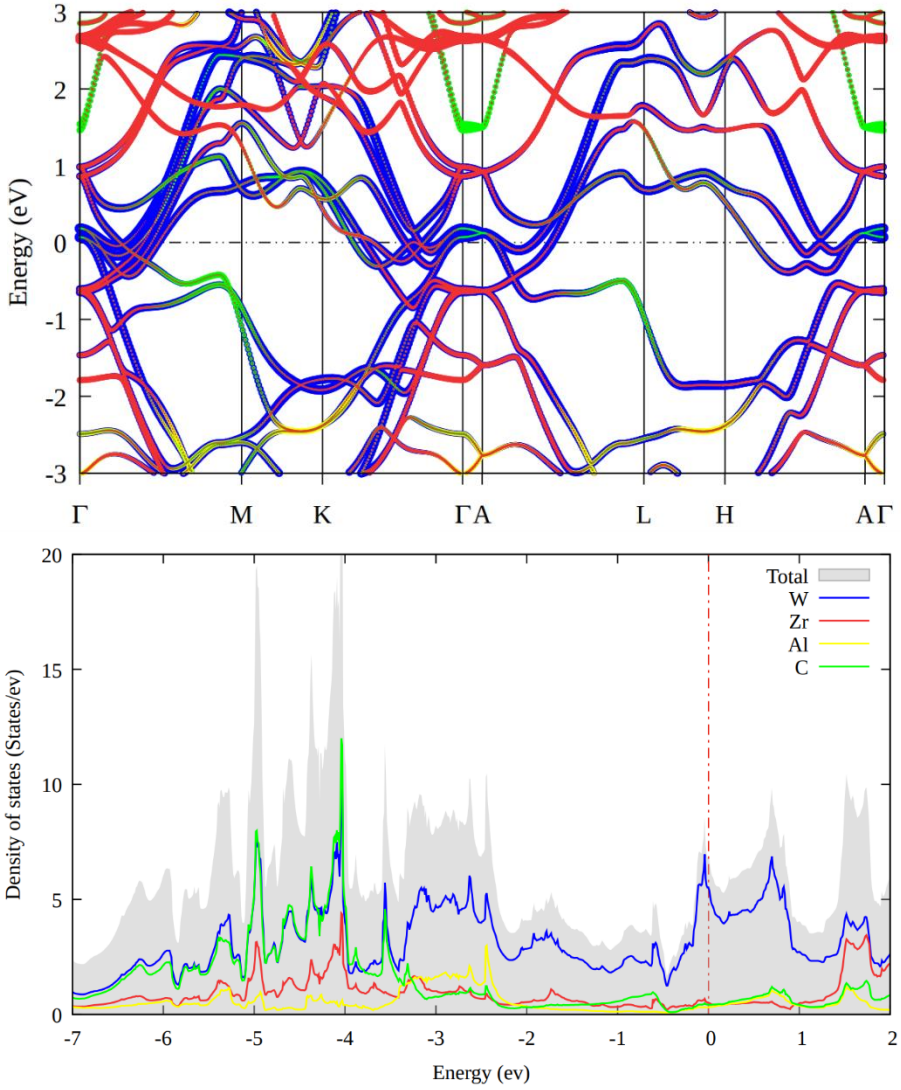
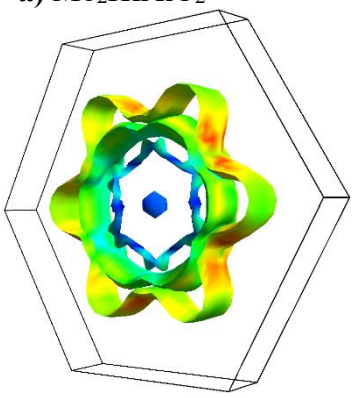
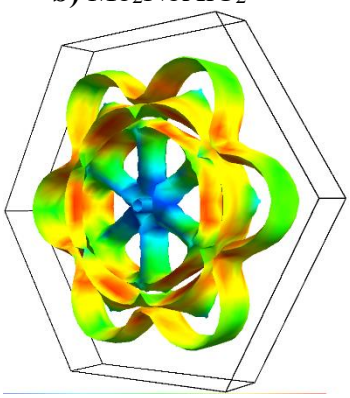



**Fig 2S.** Total and partial Band structure and density of states of **a)** $Mo_2HfAlC_2$ **b)** $Mo_2NbAlC_2$ **c)** $Mo_2ScAlC_2$ **d)** $Mo_2TaAlC_2$ **e)** $Mo_2TiAlC_2$ **f)** $Mo_2VAlC_2$ **g)** $Mo_2YAlC_2$ **h)** $Mo_2ZrAlC_2$ **i)** $Nb_2HfAlC_2$ **j)** $Nb_2ScAlC_2$ **k)** $Nb_2TaAlC_2$ **l)** $Nb_2TiAlC_2$ **m)** $Nb_2YAlC_2$ **n)** $Nb_2ZrAlC_2$ **o)** $W_2HfAlC_2$ **p)** $W_2NbAlC_2$ **q)** $W_2ScAlC_2$ **r)** $W_2TaAlC_2$ **s)** $W_2TiAlC_2$ **t)** $W_2VAlC_2$ **u)** $W_2YAlC_2$ **v)** $W_2ZrAlC_2$ (The Fermi energy is set at zero).

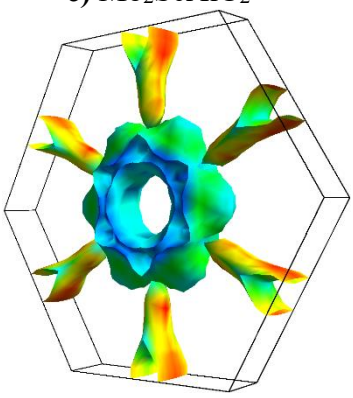
**a)** $Mo_2HfAlC_2$



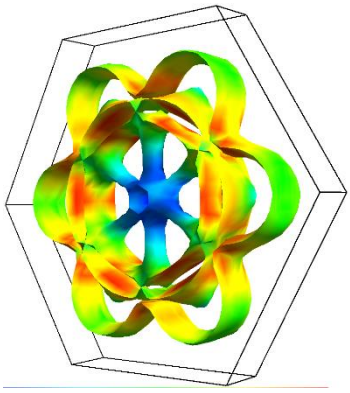
**b)** $Mo_2NbAlC_2$



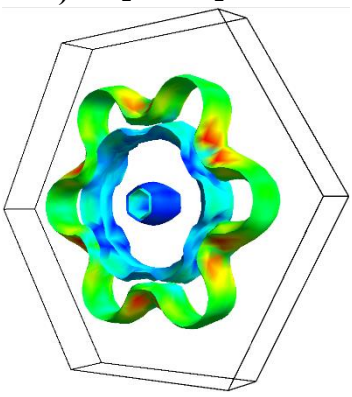
**c)** $Mo_2ScAlC_2$



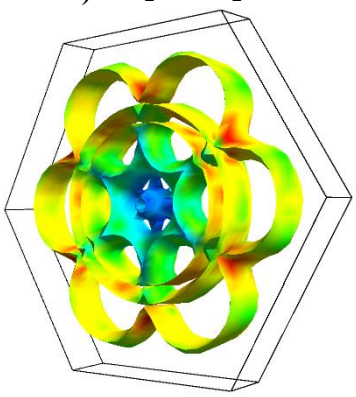
**d)** $Mo_2TaAlC_2$



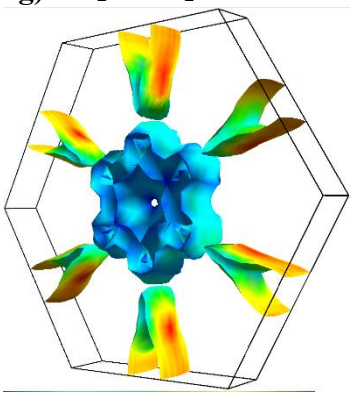
**e)** $Mo_2TiAlC_2$



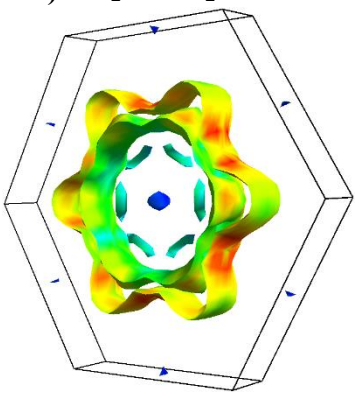
**f)** $Mo_2VAlC_2$



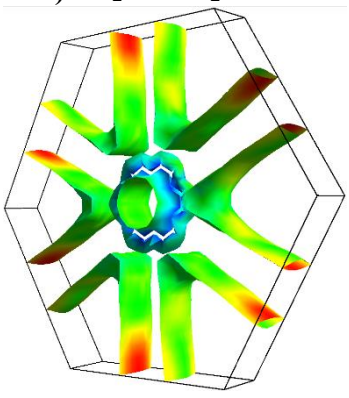
**g)** $Mo_2YAlC_2$



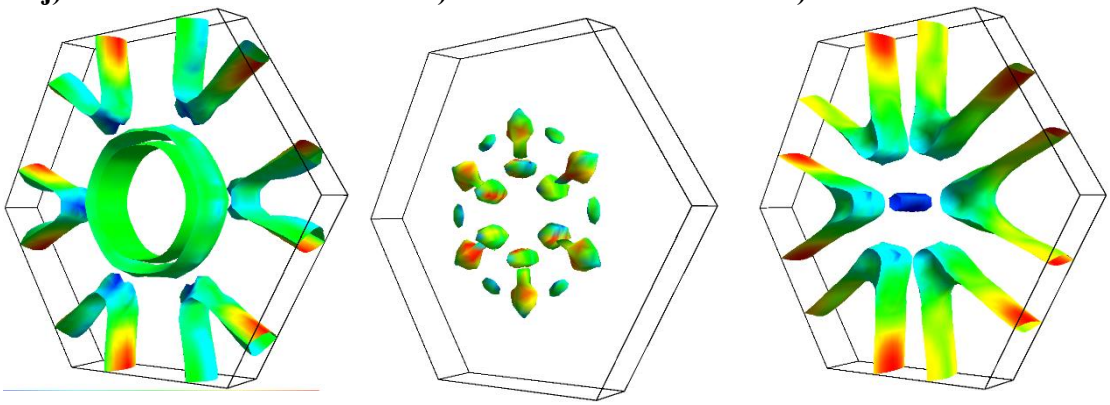
**h)** $Mo_2ZrAlC_2$



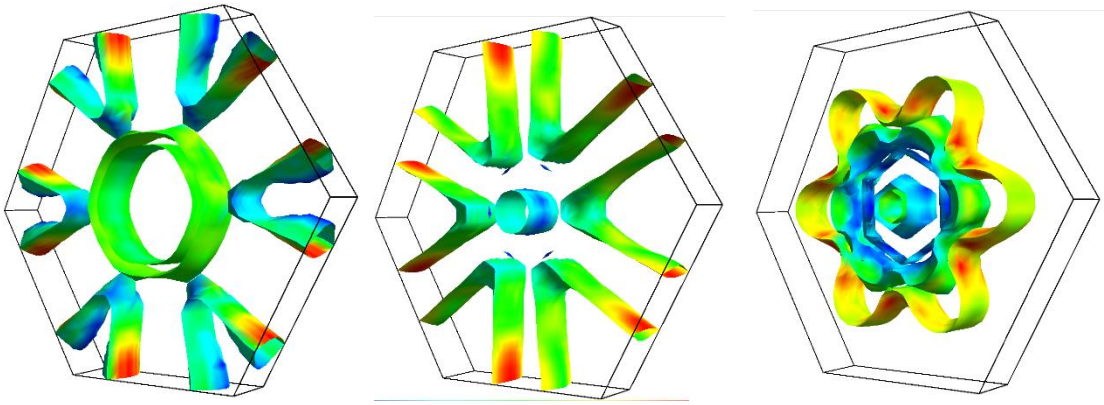
**i)** $Nb_2HfAlC_2$



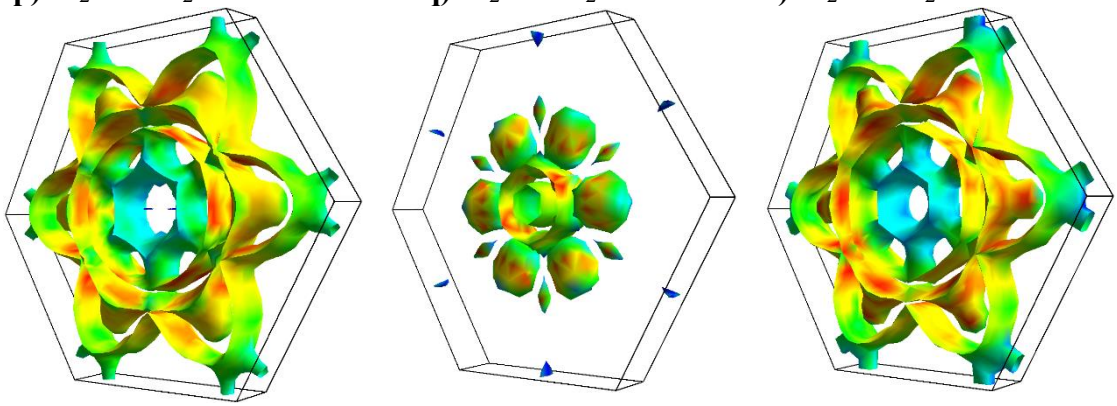
**j)** $Nb_2ScAlC_2$ **k)** $Nb_2TaAlC_2$ **l)** $Nb_2TiAlC_2$



**m)** $Nb_2YAlC_2$ **n)** $Nb_2ZrAlC_2$ **o)** $W_2HfAlC_2$



**p)** $W_2NbAlC_2$ **q)** $W_2ScAlC_2$ **r)** $W_2TaAlC_2$



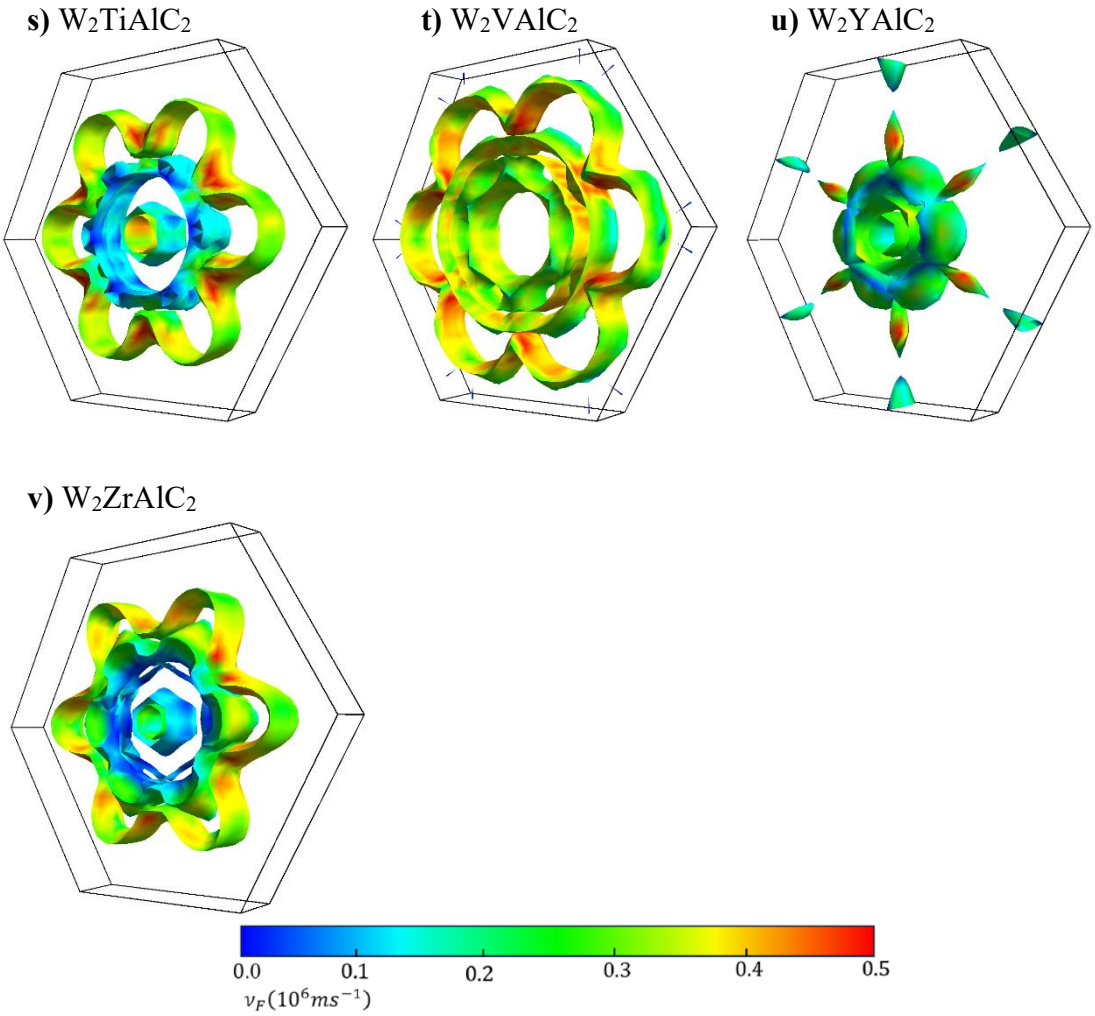
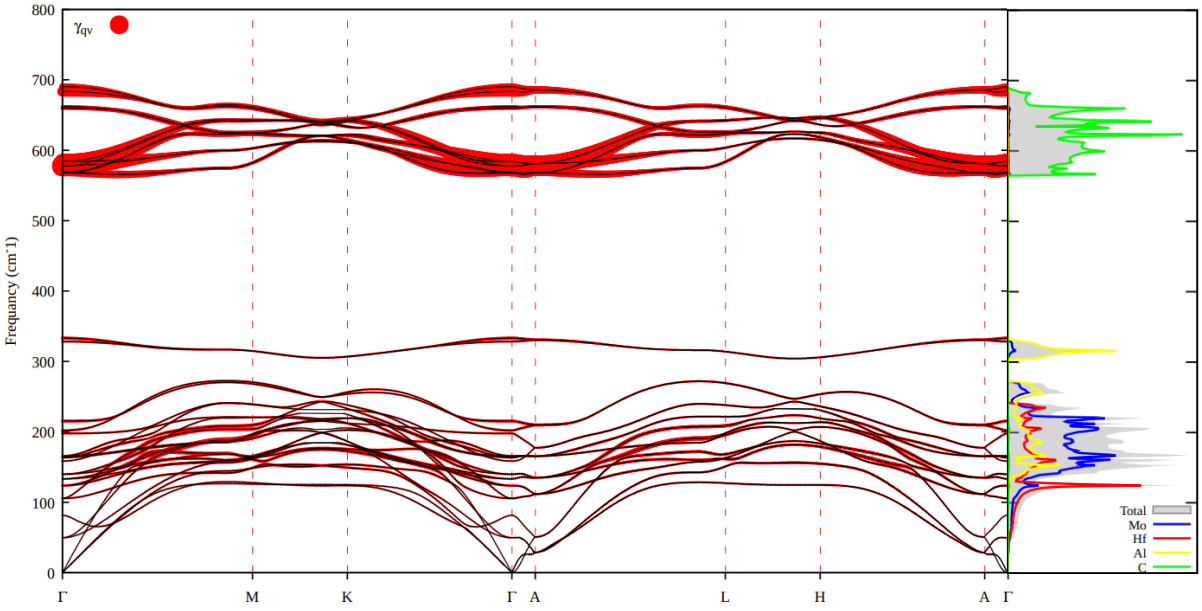
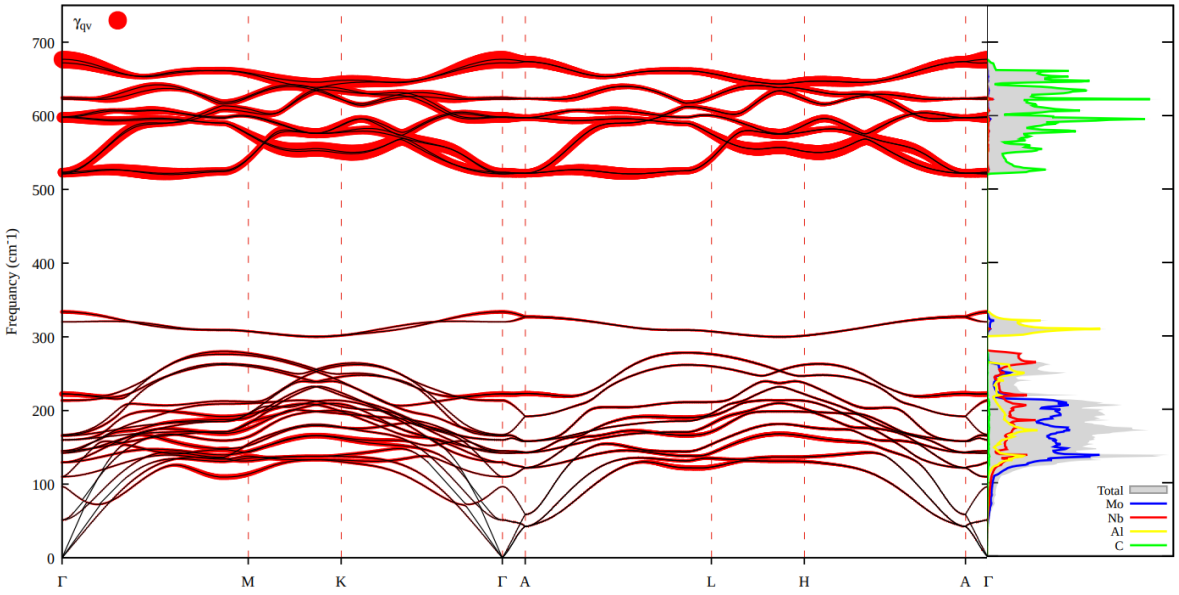



**Fig 3S**. 3D Fermi surface combined with color drawn to indicate the relative Fermi velocity $\nu_F$ for **a)** $Mo_2HfAlC_2$ **b)** $Mo_2NbAlC_2$ **c)** $Mo_2ScAlC_2$ **d)** $Mo_2TaAlC_2$ **e)** $Mo_2TiAlC_2$ **f)** $Mo_2VAlC_2$ **g)** $Mo_2YAlC_2$ **h)** $Mo_2ZrAlC_2$ **i)** $Nb_2HfAlC_2$ **j)** $Nb_2ScAlC_2$ **k)** $Nb_2TaAlC_2$ **l)** $Nb_2TiAlC_2$ **m)** $Nb_2YAlC_2$ **n)** $Nb_2ZrAlC_2$ **o)** $W_2HfAlC_2$ **p)** $W_2NbAlC_2$ **q)** $W_2ScAlC_2$ **r)** $W_2TaAlC_2$ **s)** $W_2TiAlC_2$ **t)** $W_2VAlC_2$ **u)** $W_2YAlC_2$ **v)** $W_2ZrAlC_2$.

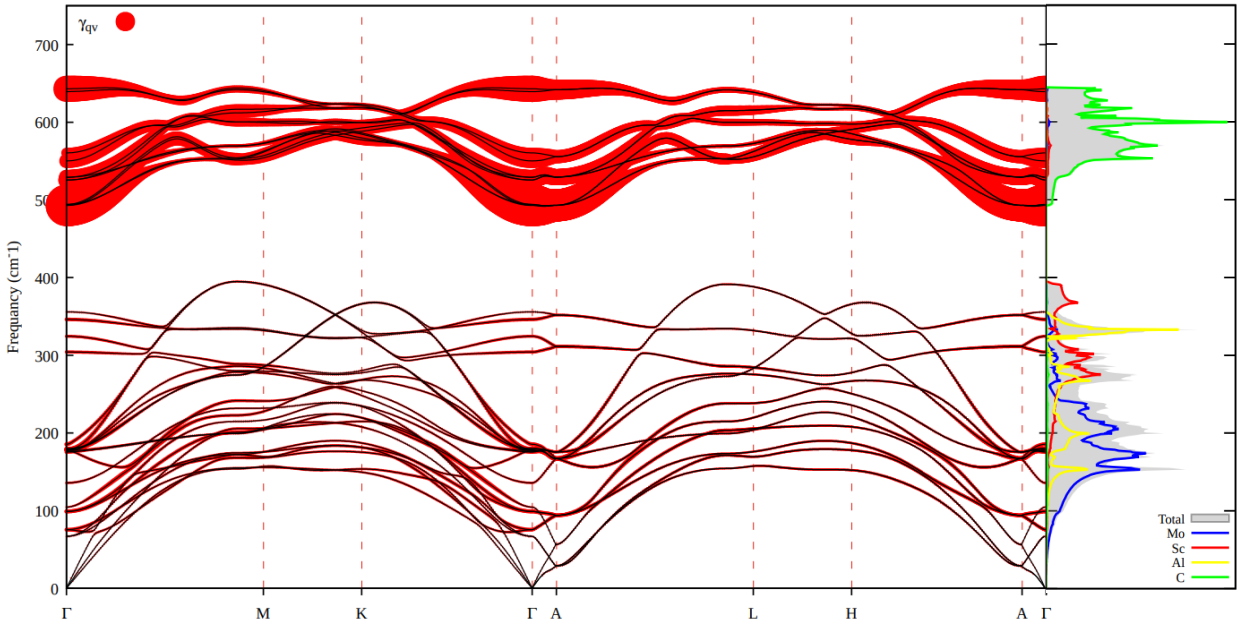
**a)** $Mo_2HfAlC_2$



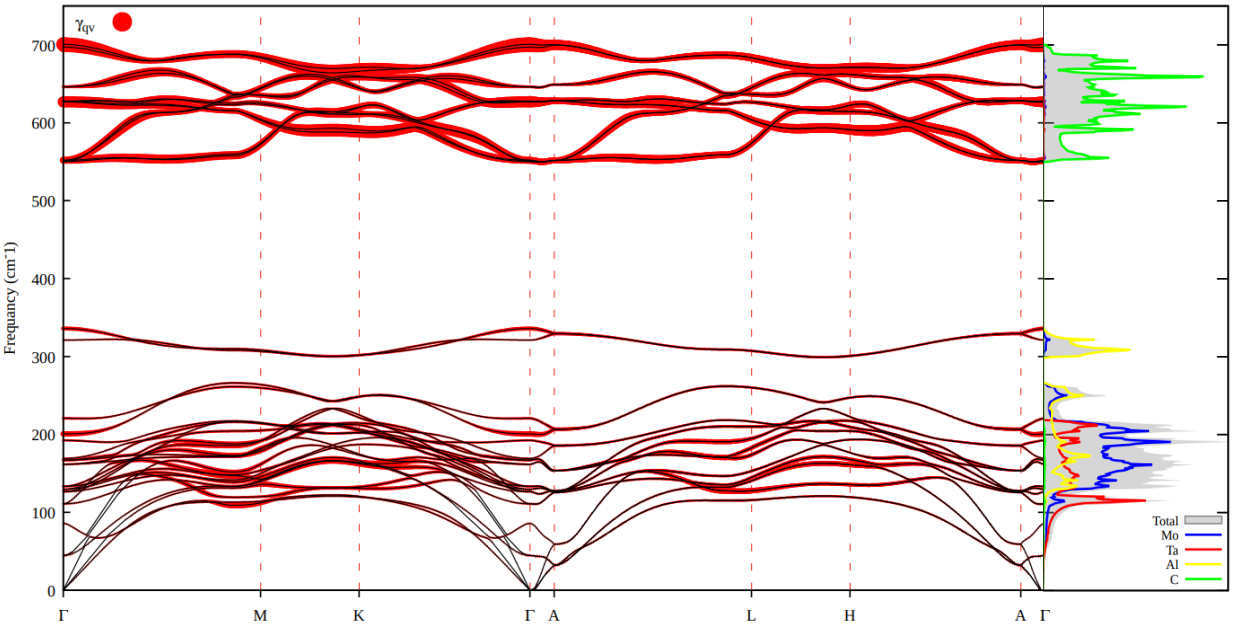

**b)** $Mo_2NbAlC_2$



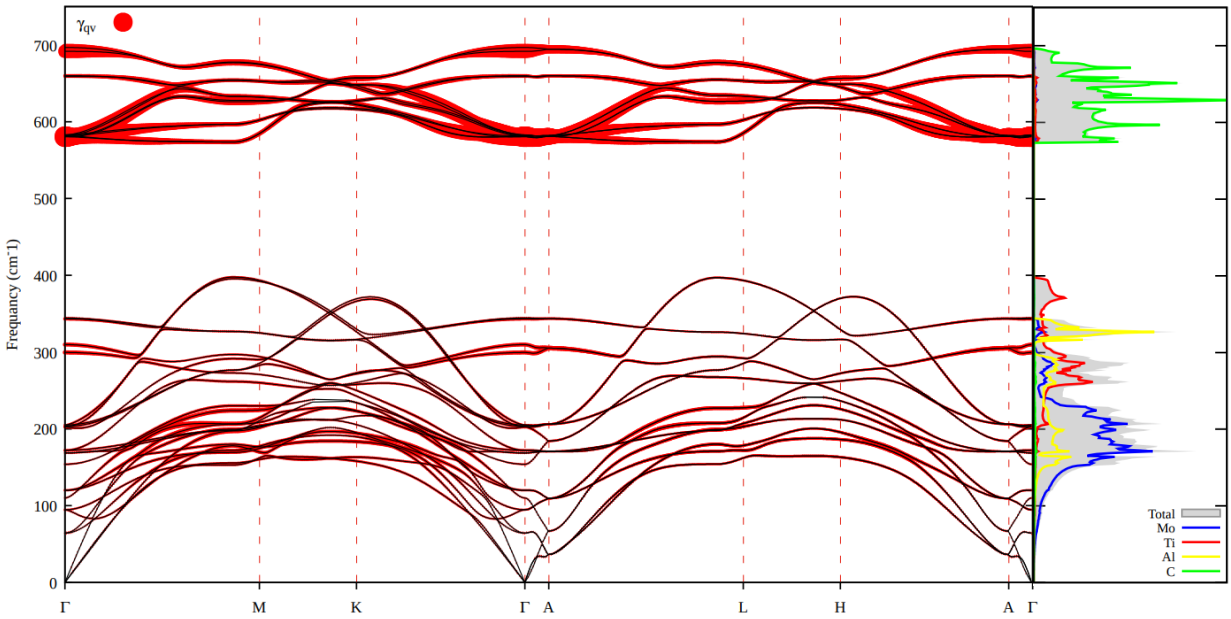
**c)** $Mo_2ScAlC_2$



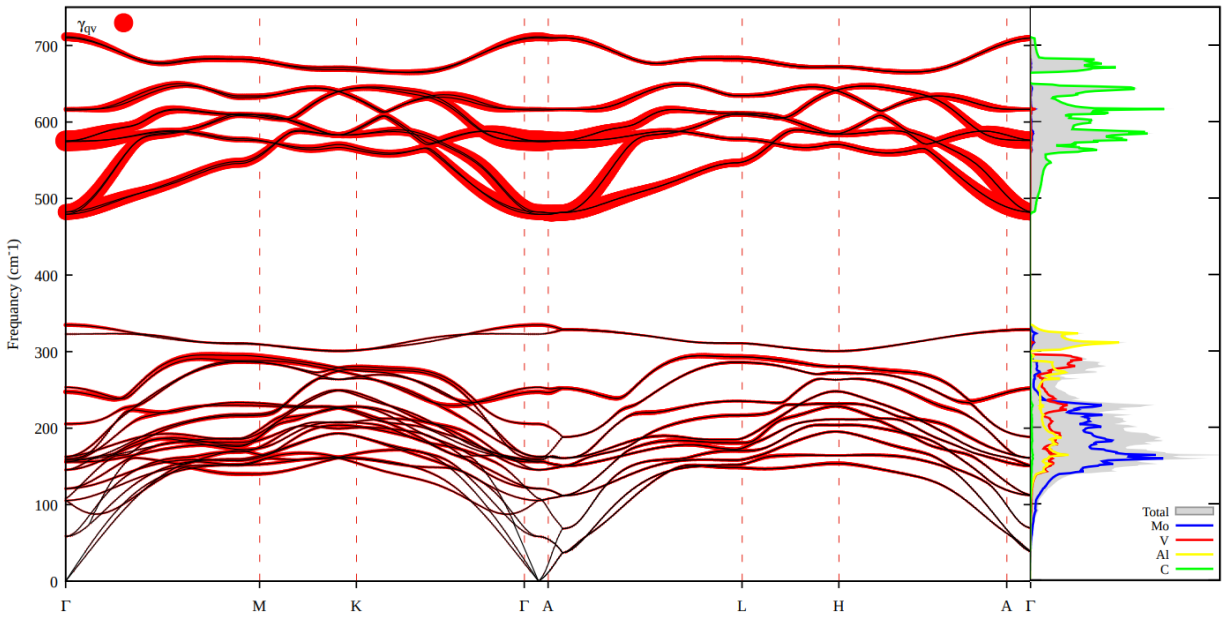

**d)** $Mo_2TaAlC_2$



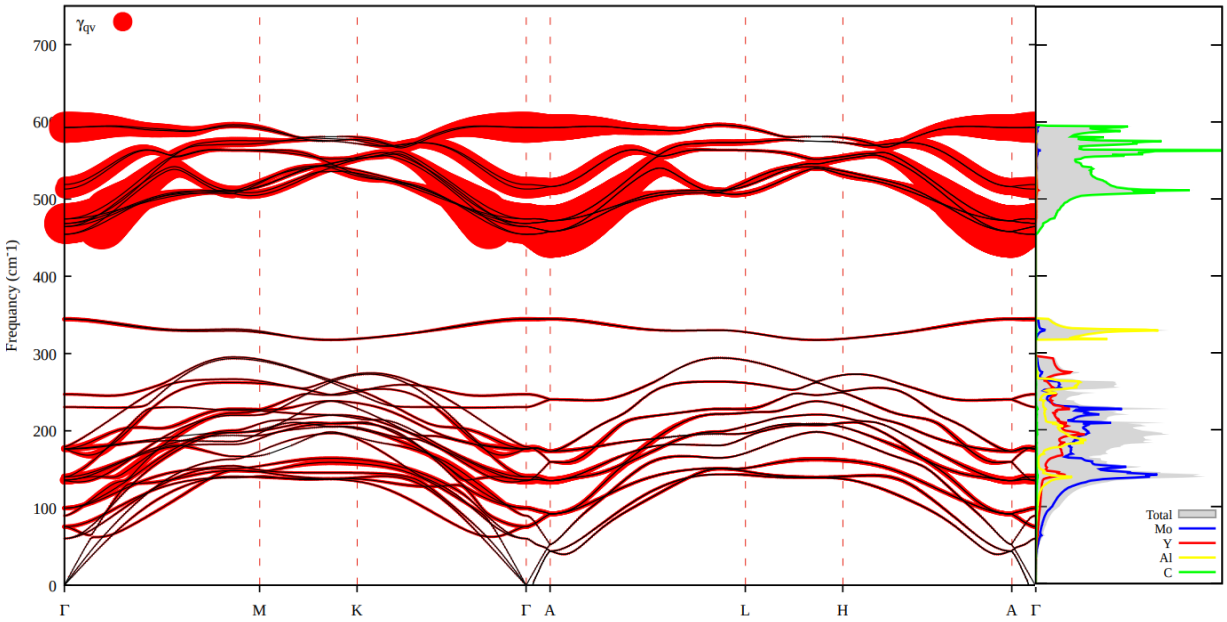
**e)** $Mo_2TiAlC_2$



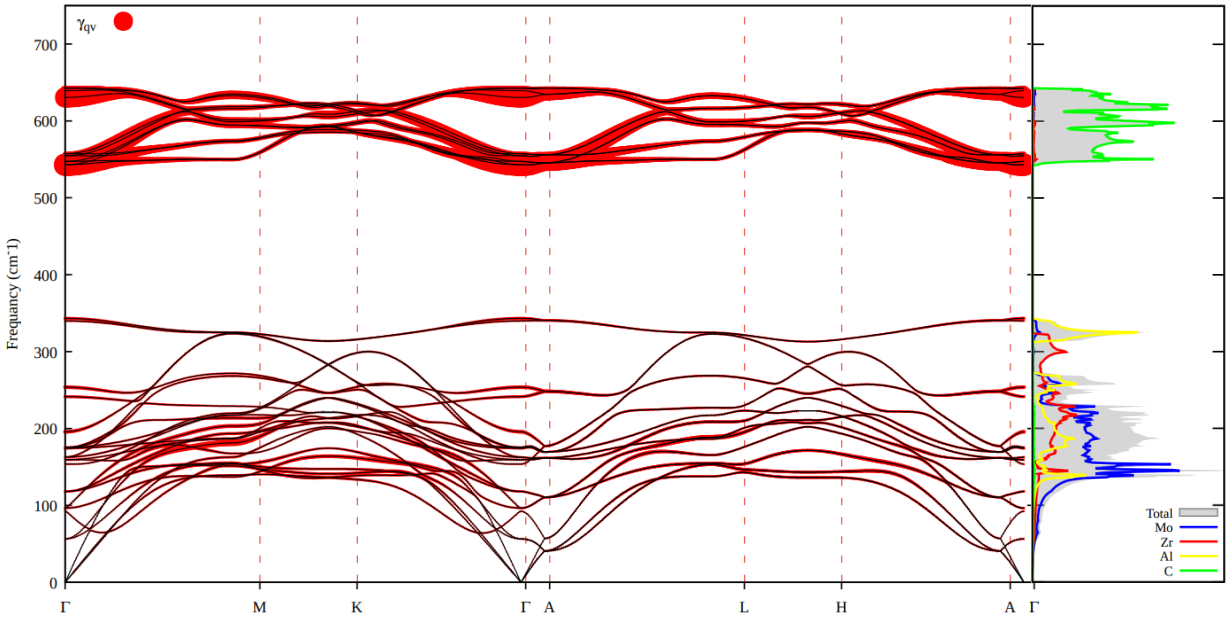

**f)** $Mo_2VAlC_2$



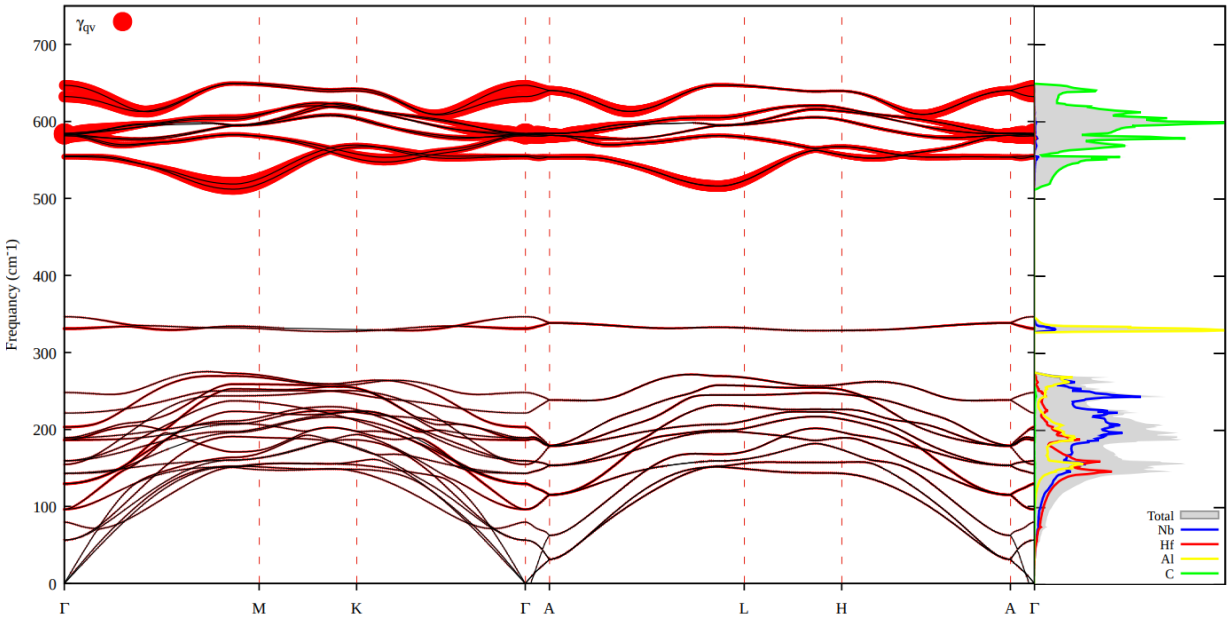
**g)** $Mo_2YAlC_2$



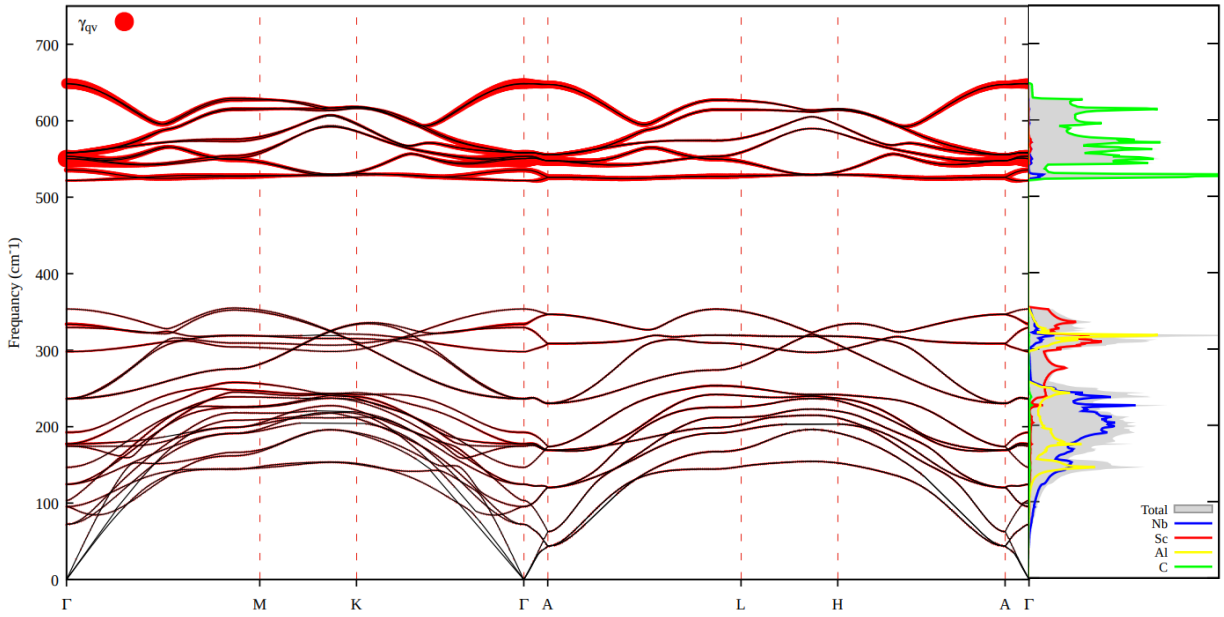

**h)** $Mo_2ZrAlC_2$



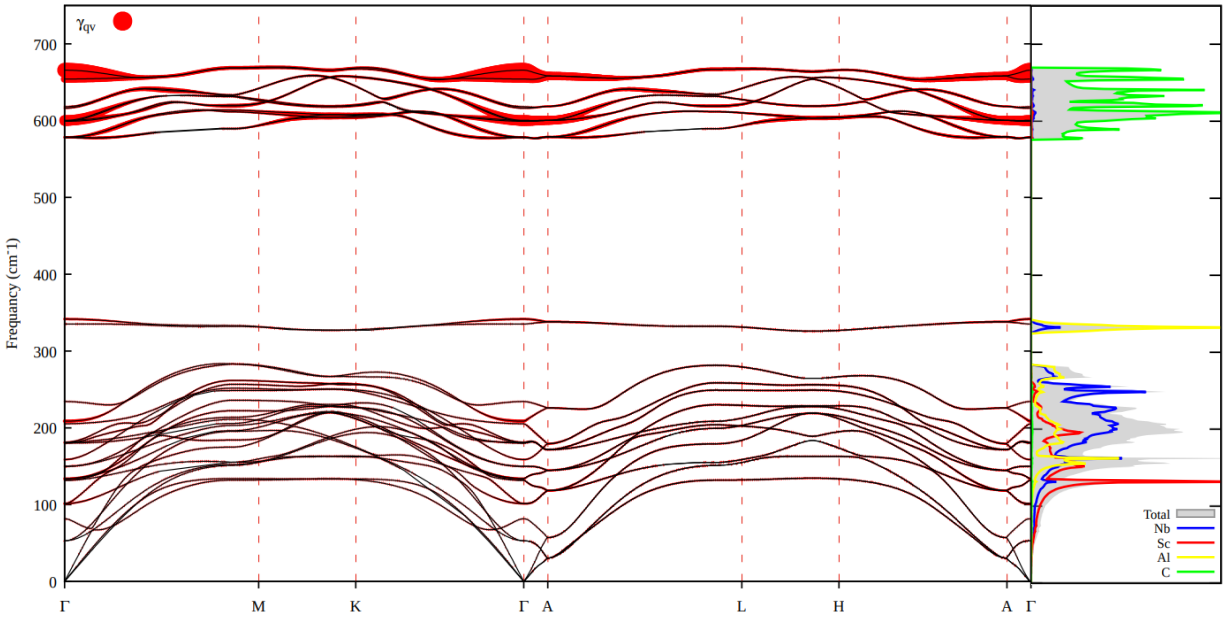
**i)** $Nb_2HfAlC_2$



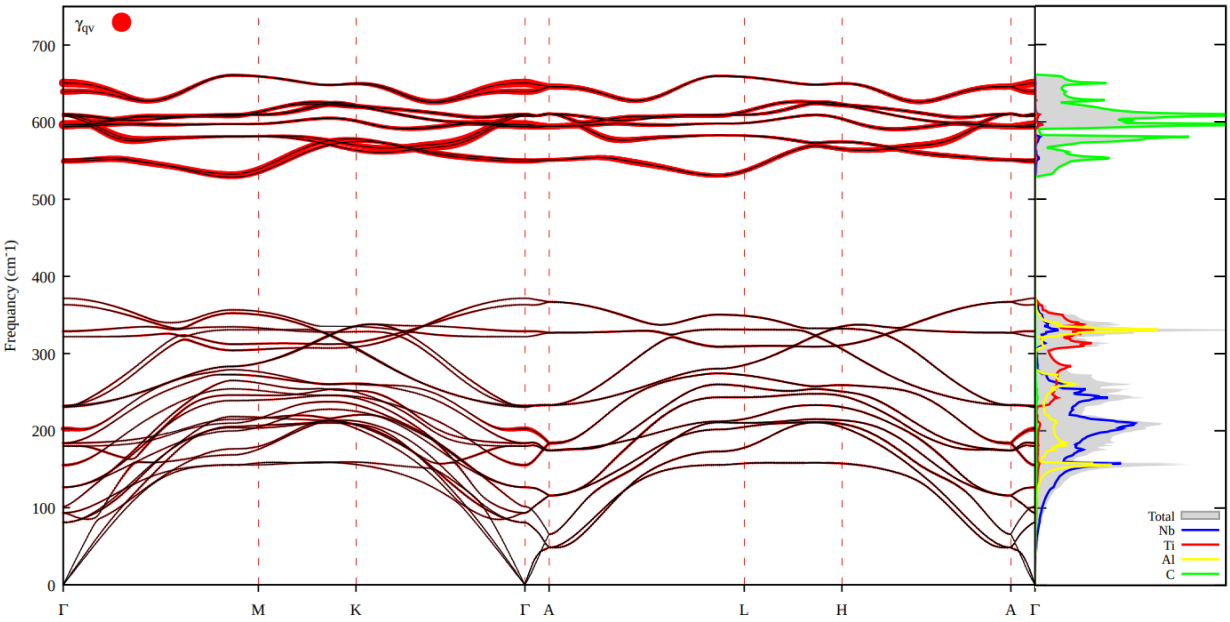

**j)** $Nb_2ScAlC_2$



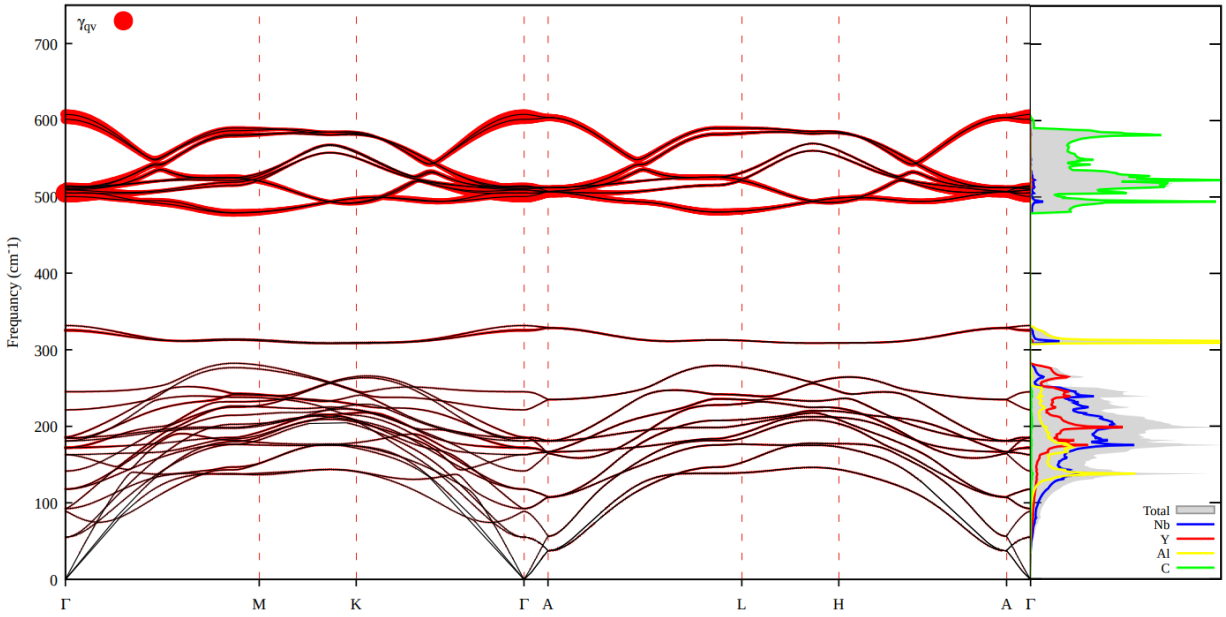
**k)** $Nb_2TaAlC_2$



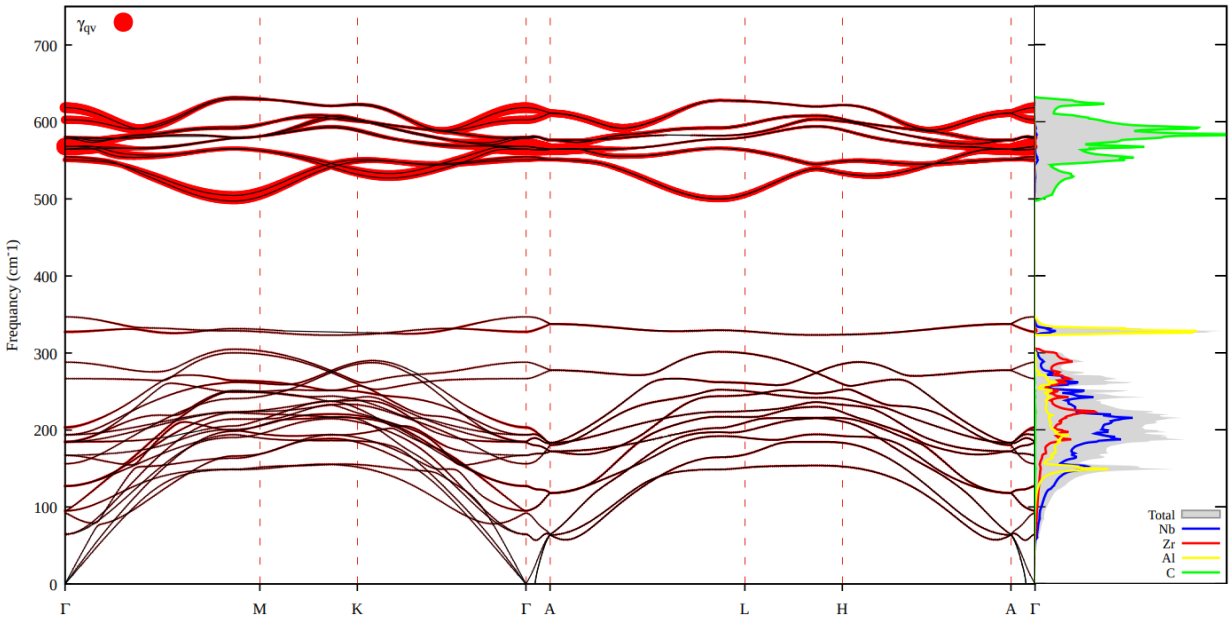

**l)** $Nb_2TiAlC_2$



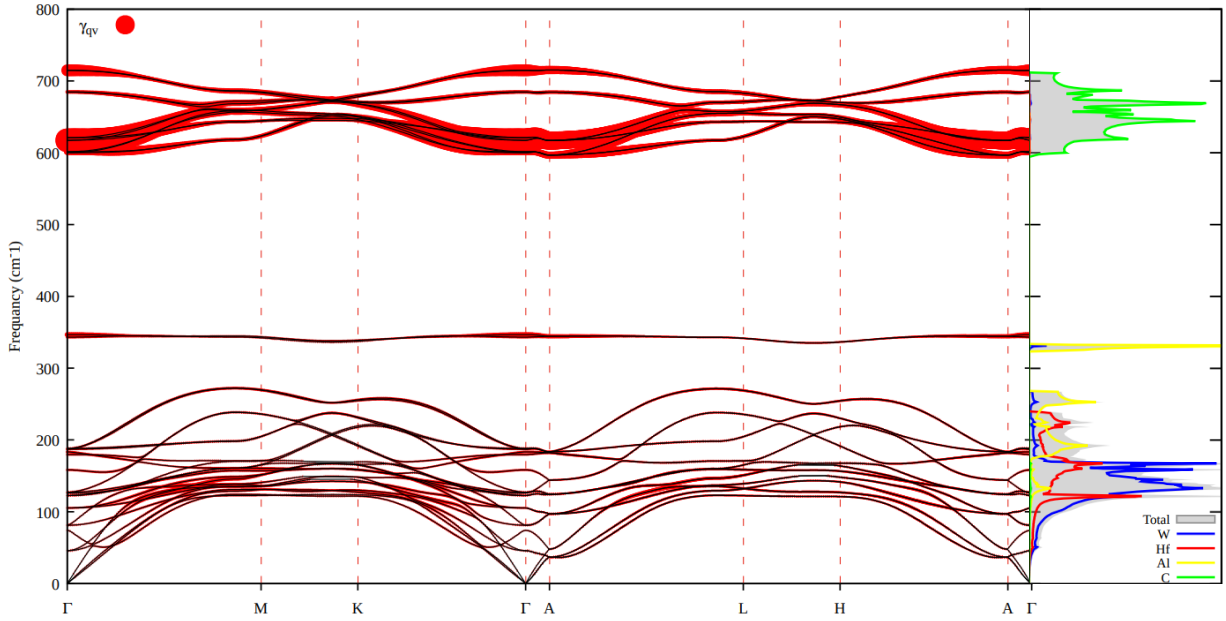
**m)** $Nb_2YAlC_2$


γqv
Frequancy (cm-1)
700
600
500
400
300
200
100
0
Γ
M
K
Γ
A
L
H
A
Γ
Total
Nb
Y
Al
C

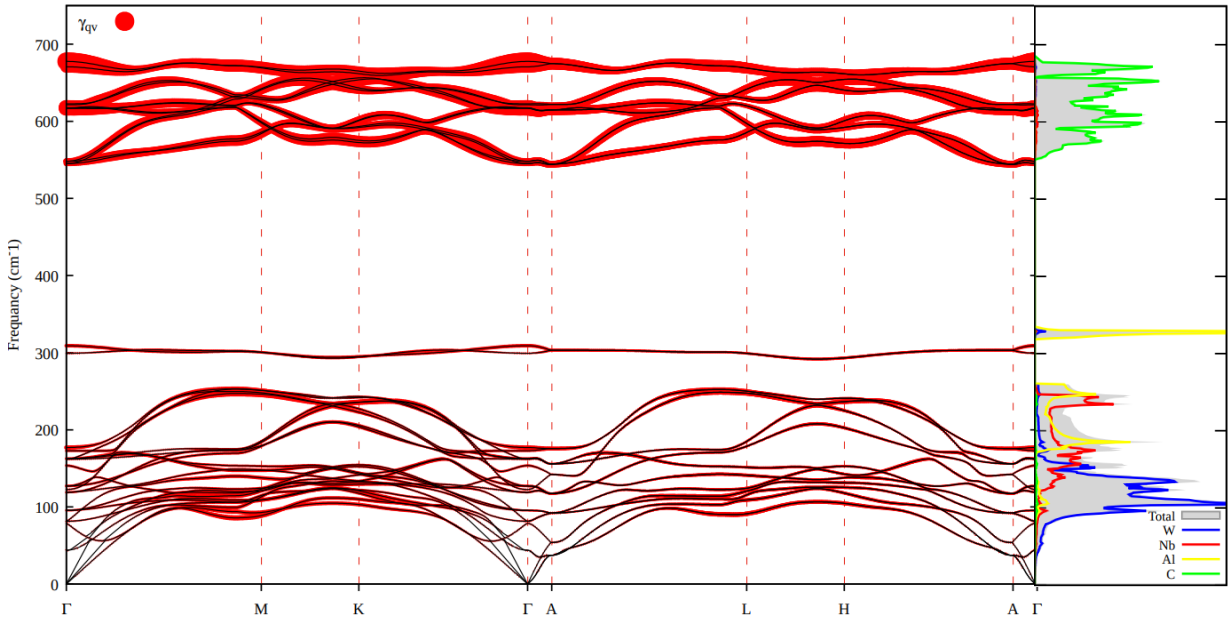
**n)** $Nb_2ZrAlC_2$



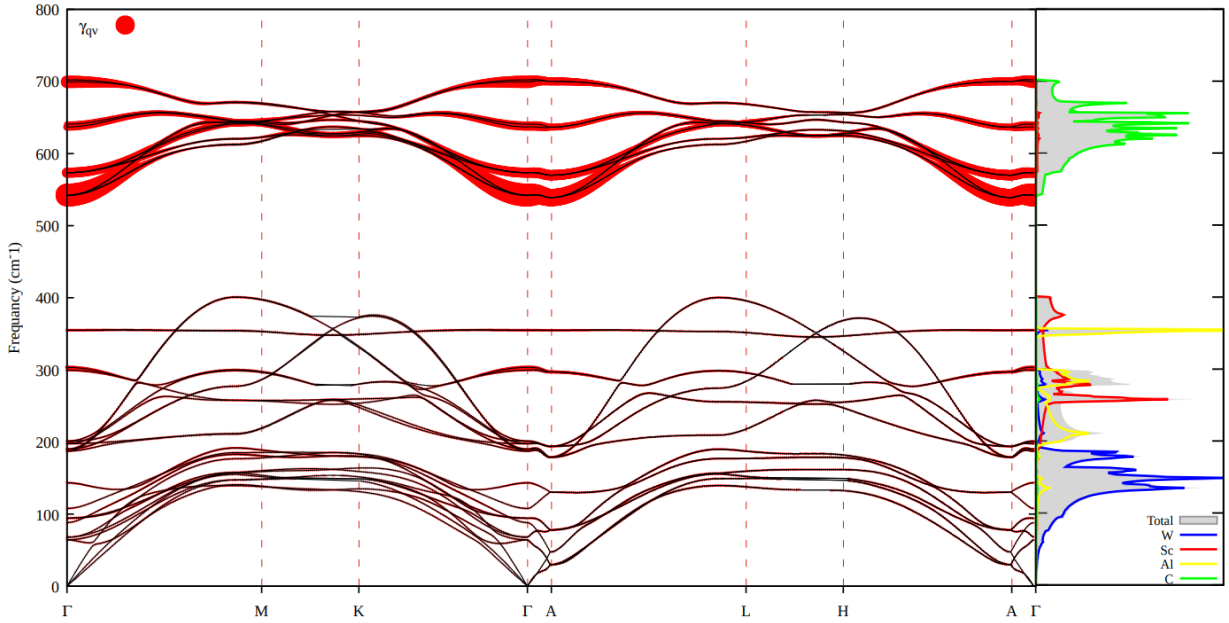

**o)** $W_2HfAlC_2$



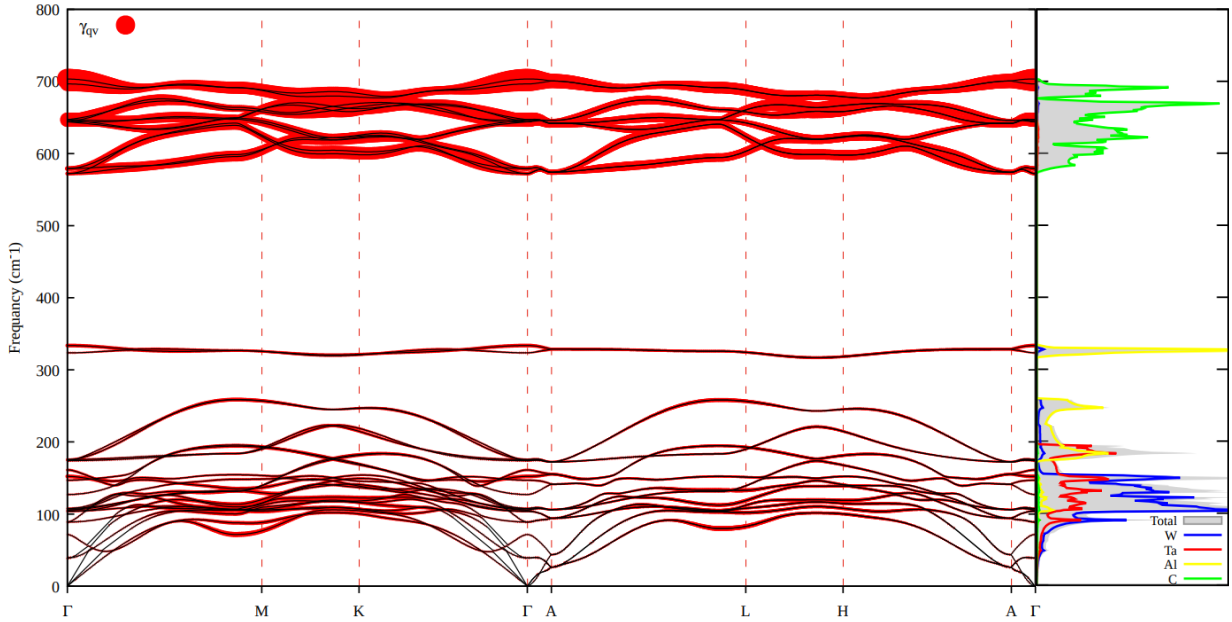
**p)** $W_2NbAlC_2$



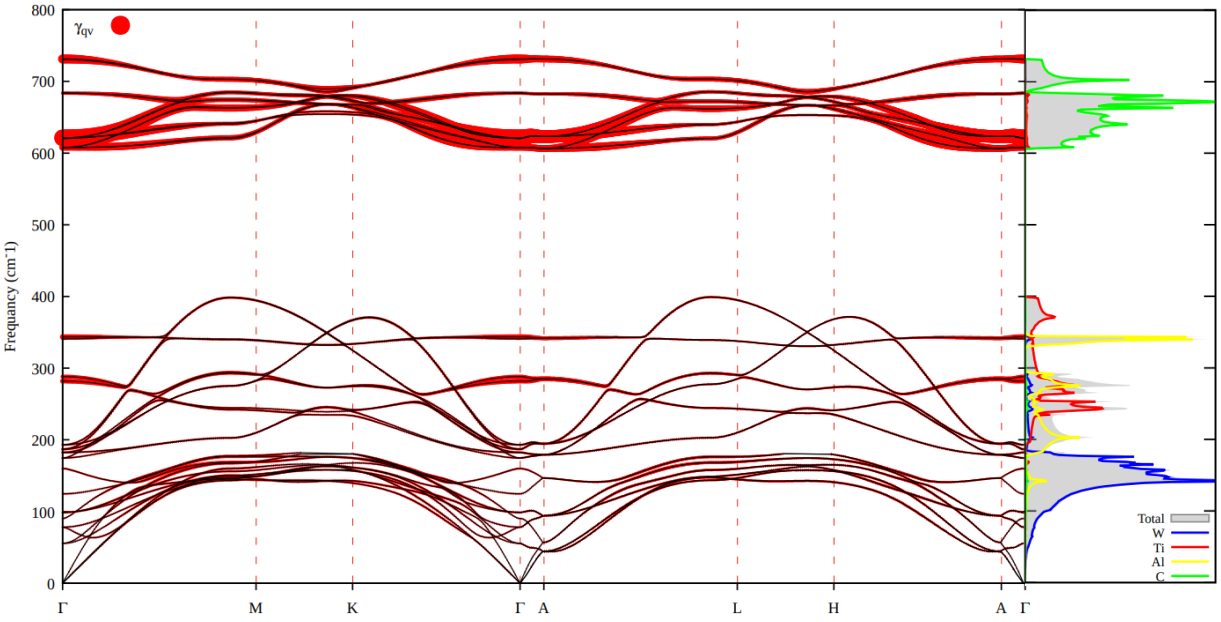

**q)** $W_2ScAlC_2$



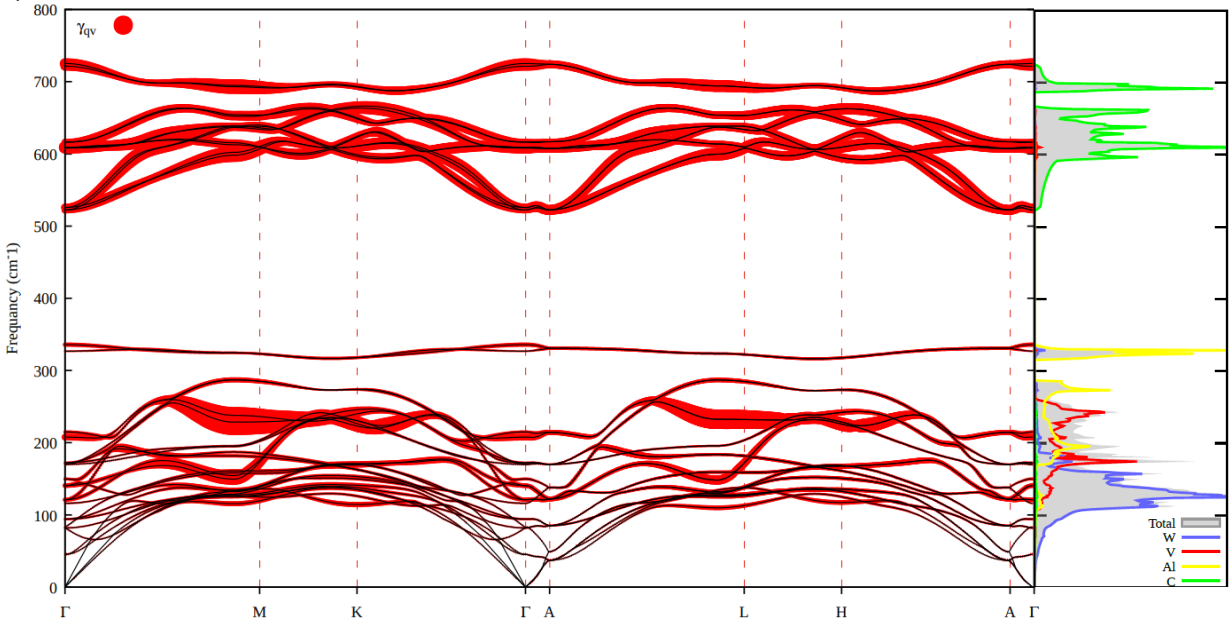
**r)** $W_2TaAlC2$



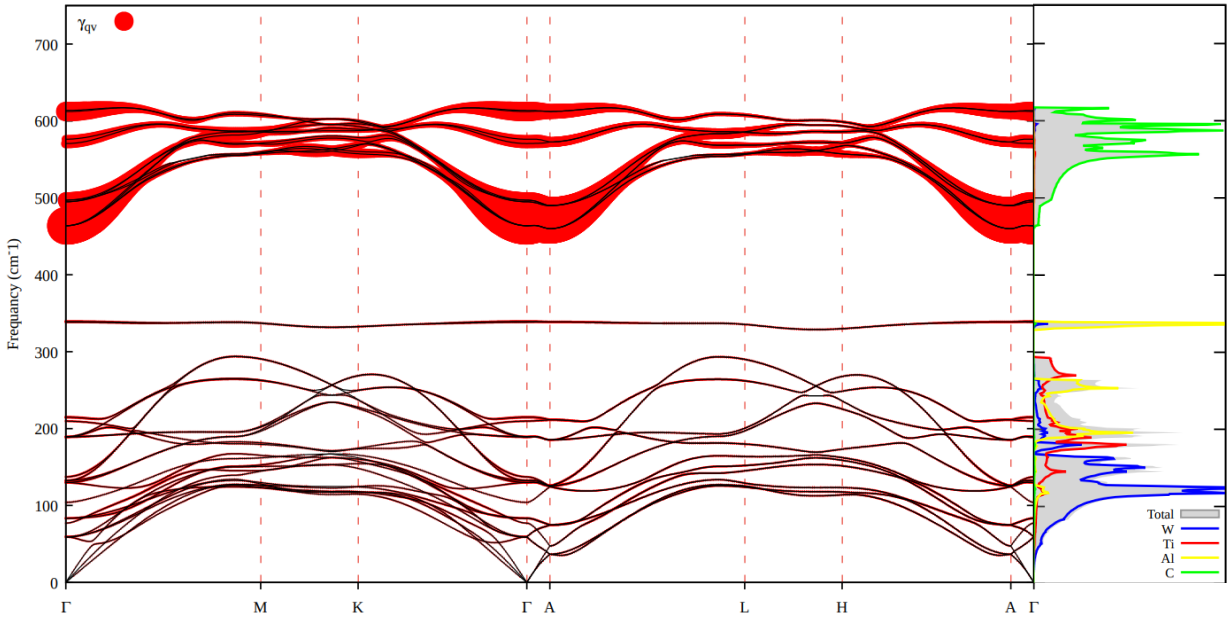

**s)** $W_2TiAlC_2$



**t)** $W_2VAlC_2$




**u)** $W_2YAlC_2$



**v)** $W_2ZrAlC_2$

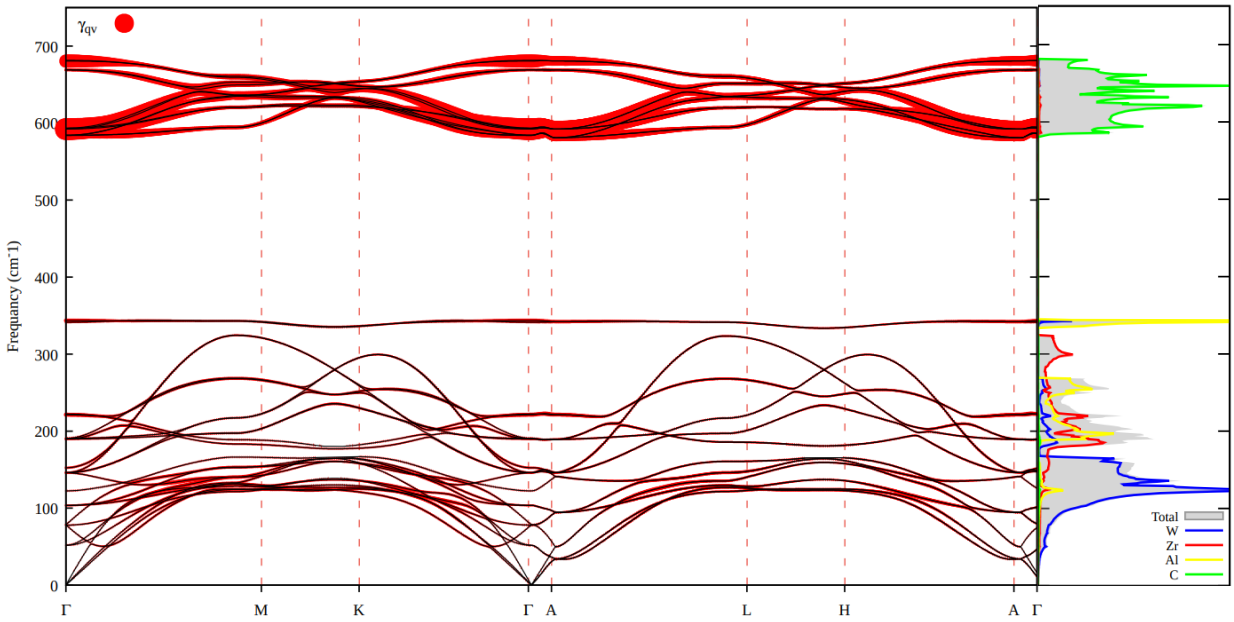
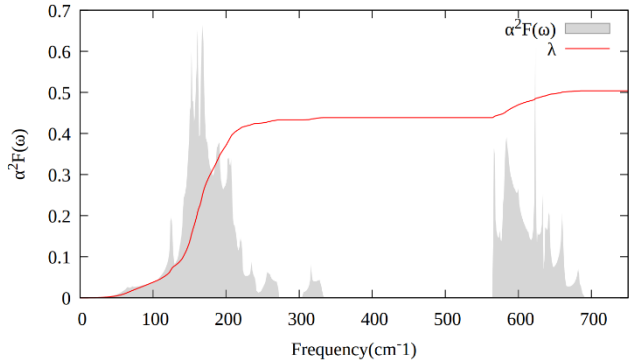
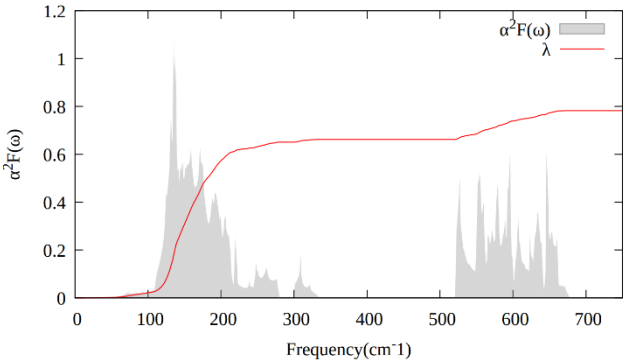



**Fig 4S.** Phonon dispersion curves, total and partial vibrational density of states for **a)** $Mo_2HfAlC_2$ **b)** $Mo_2NbAlC_2$ **c)** $Mo_2ScAlC_2$ **d)** $Mo_2TaAlC_2$ **e)** $Mo_2TiAlC_2$ **f)** $Mo_2VAlC_2$ **g)** $Mo_2YAlC_2$ **h)** $Mo_2ZrAlC_2$ **i)** $Nb_2HfAlC_2$ **j)** $Nb_2ScAlC_2$ **k)** $Nb_2TaAlC_2$ **l)** $Nb_2TiAlC_2$ **m)** $Nb_2YAlC_2$ **n)** $Nb_2ZrAlC_2$ **o)** $W_2HfAlC_2$ **p)** $W_2NbAlC_2$ **q)** $W_2ScAlC_2$ **r)** $W_2TaAlC_2$ **s)** $W_2TiAlC_2$ **t)** $W_2VAlC_2$ **u)** $W_2YAlC_2$ **v)** $W_2ZrAlC_2$ (Phonon dispersion curves are weighted by phonon linewidth ($\gamma_{qv}$)).

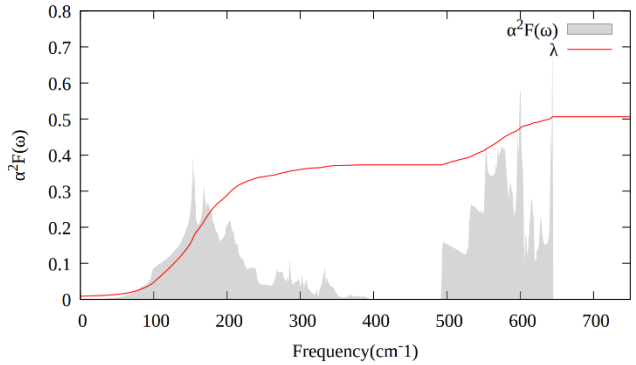
**a)** $Mo_2HfAlC_2$



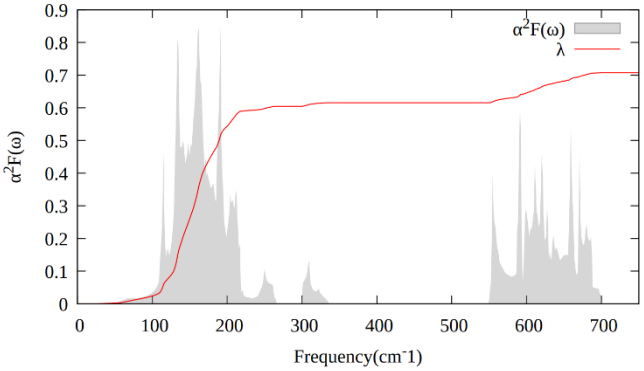

**b)** $Mo_2NbAlC_2$



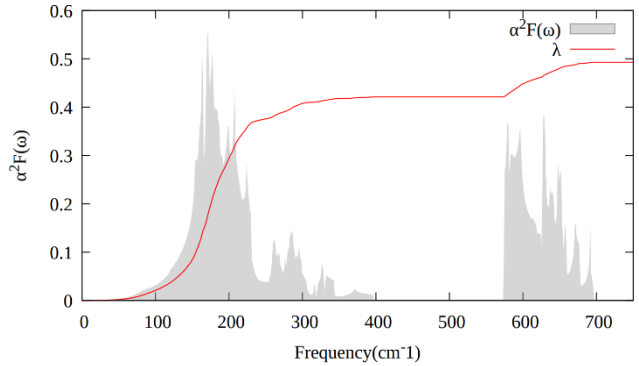

**c)** $Mo_2ScAlC_2$



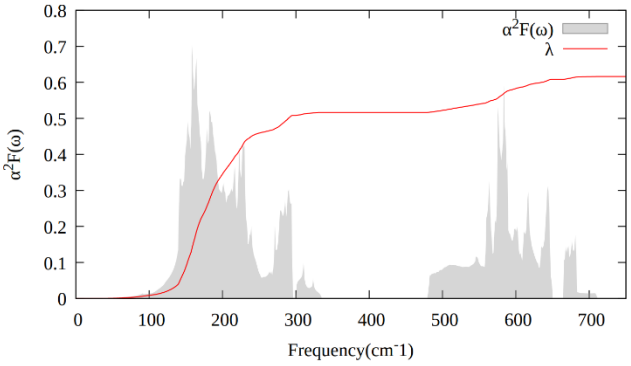

**d)** $Mo_2TaAlC_2$



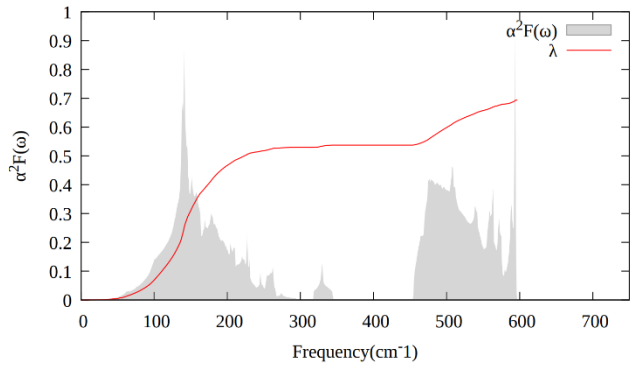

**e)** $Mo_2TiAlC_2$



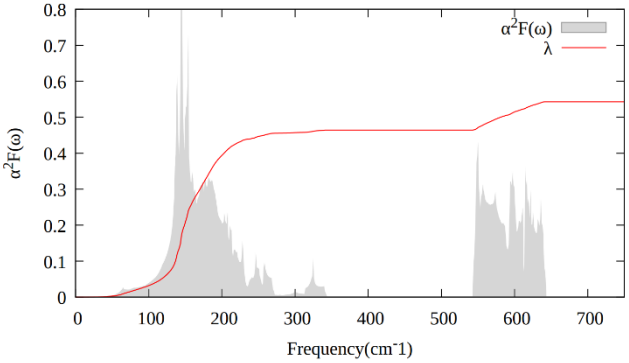

**f)** $Mo_2VAlC_2$



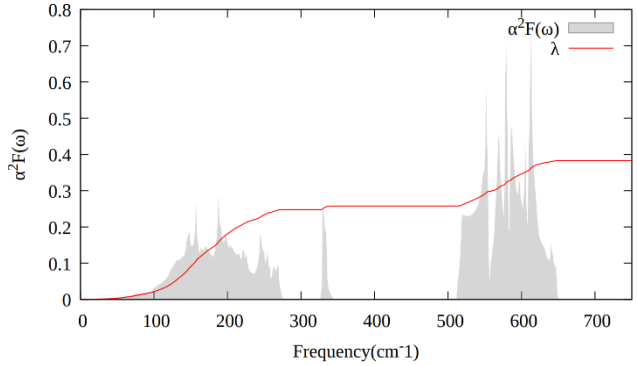
**g)** $Mo_2YAlC_2$



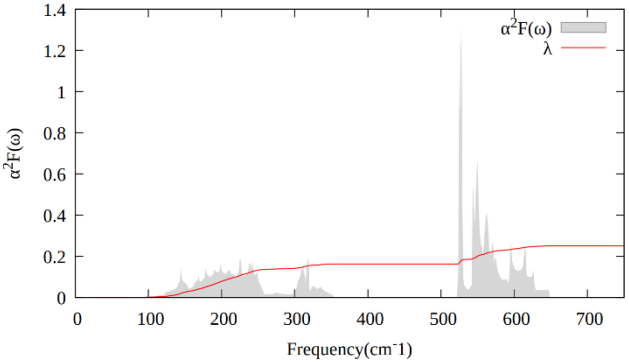

**h)** $Mo_2ZrAlC_2$



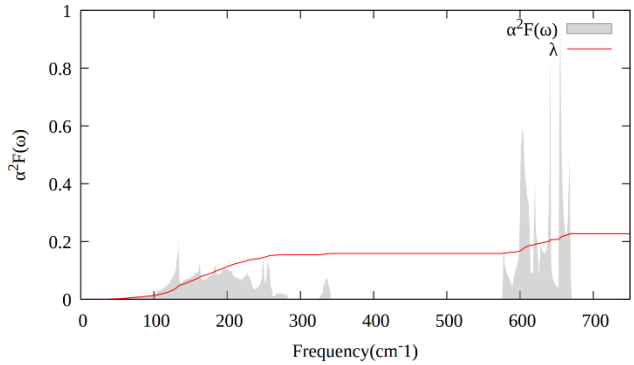

**i)** $Nb_2HfAlC_2$



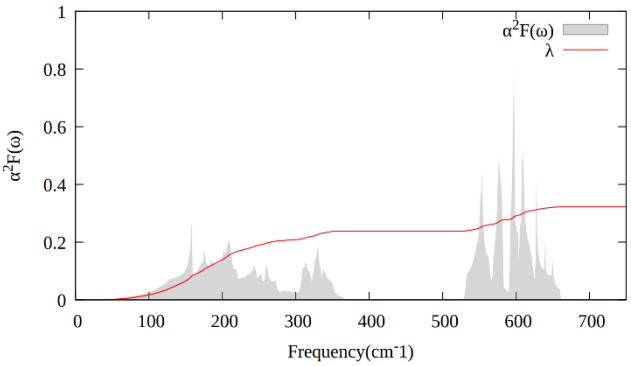

**j)** $Nb_2ScAlC_2$



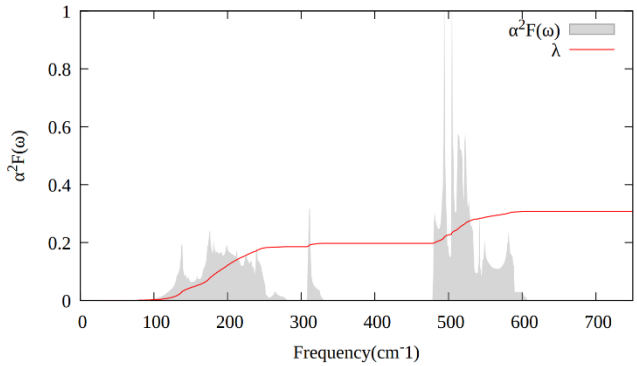

**k)** $Nb_2TaAlC_2$



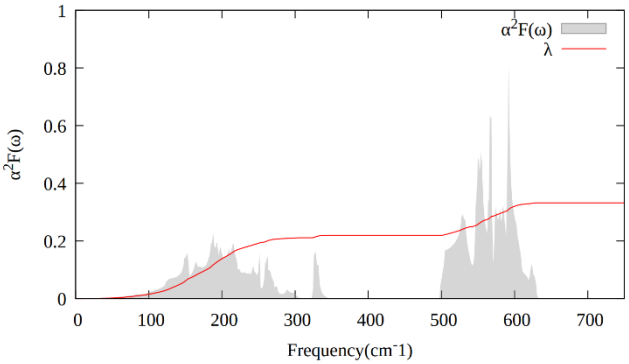

**l)** $Nb_2TiAlC_2$



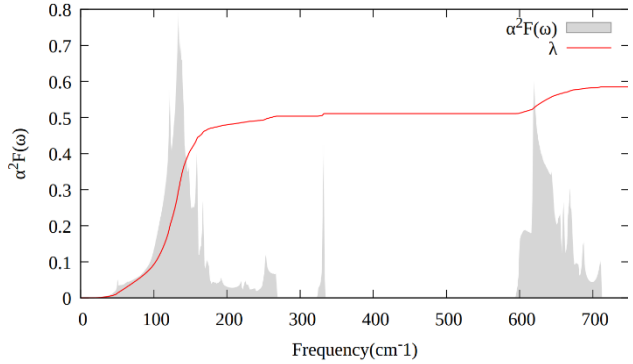
**m)** $Nb_2YAlC_2$



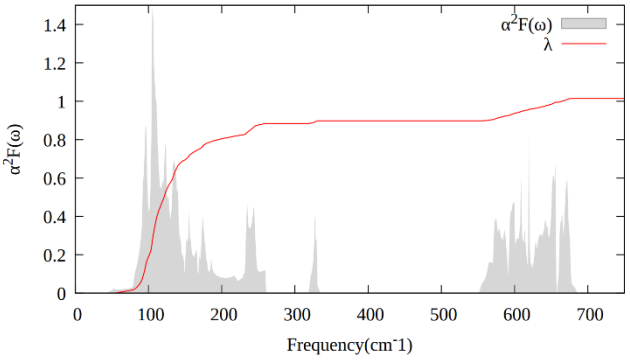

**n)** $Nb_2ZrAlC_2$



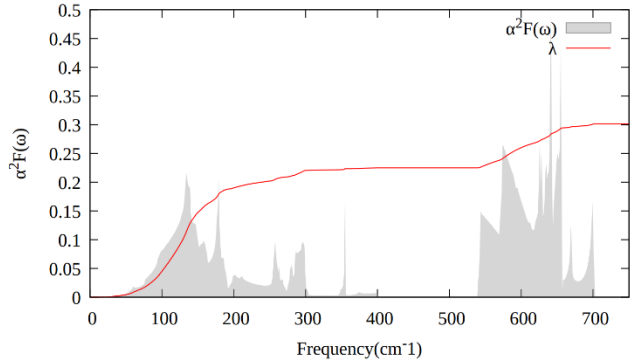

**o)** $W_2HfAlC_2$



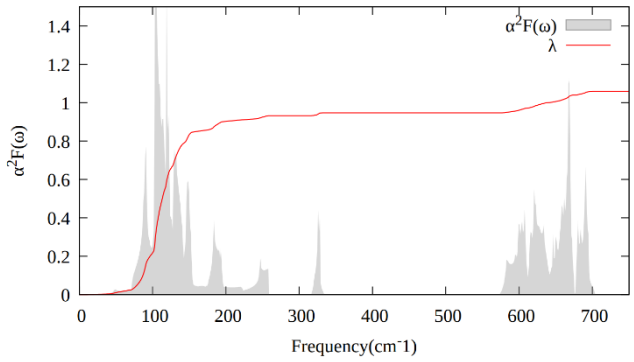

**p)** $W_2NbAlC_2$



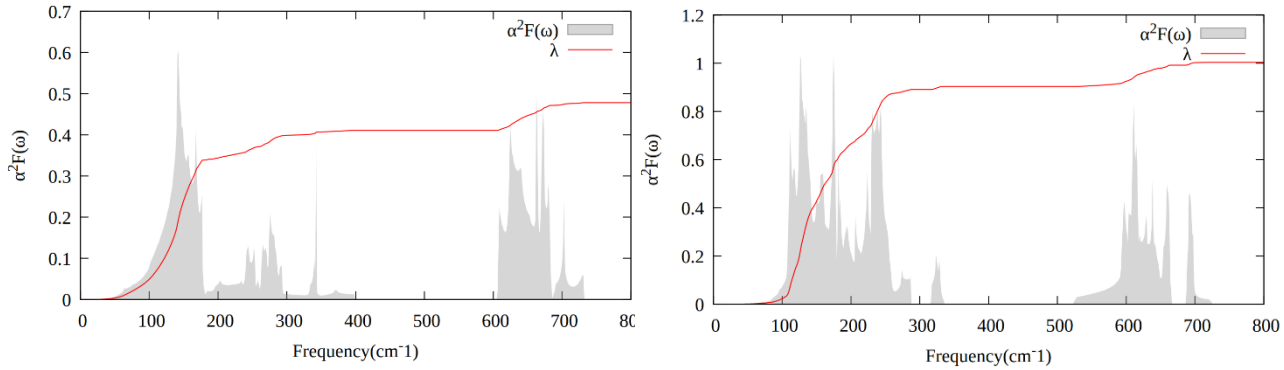

**q)** $W_2ScAlC_2$




**r)** $W_2TaAlC_2$



**s)** $W_2TiAlC_2$ **t)** $W_2VAlC_2$




**u)** $W_2YAlC_2$ **v)** $W_2ZrAlC_{2s}$

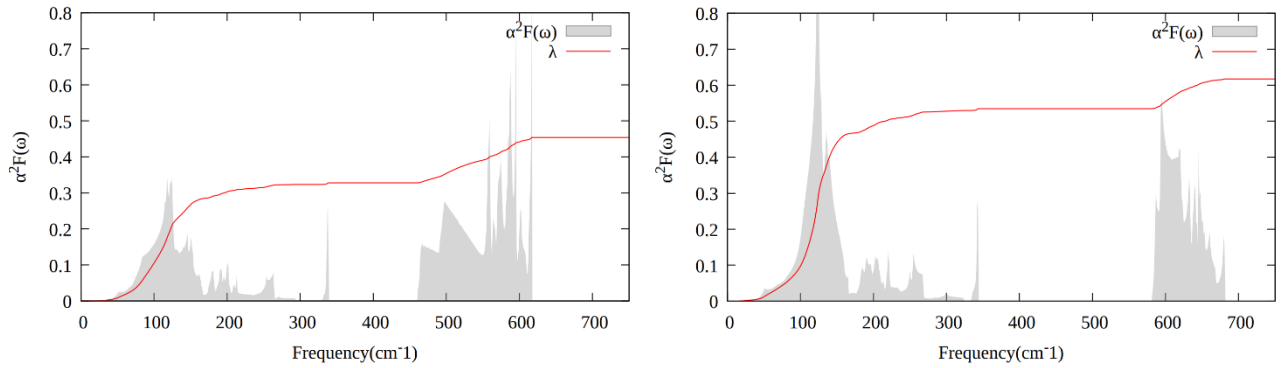


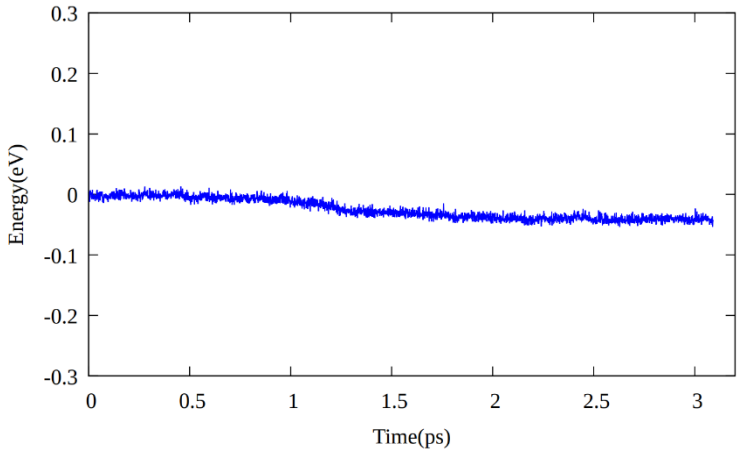

**Fig 5S.** Eliashberg spectral function $\alpha^2F(\omega)$ for **a)** $Mo_2HfAlC_2$ **b)** $Mo_2NbAlC_2$ **c)** $Mo_2ScAlC_2$ **d)** $Mo_2TaAlC_2$ **e)** $Mo_2TiAlC_2$ **f)** $Mo_2VAlC_2$ **g)** $Mo_2YAlC_2$ **h)** $Mo_2ZrAlC_2$ **i)** $Nb_2HfAlC_2$ **j)** $Nb_2ScAlC_2$ **k)** $Nb_2TaAlC_2$ **l)** $Nb_2TiAlC_2$ **m)** $Nb_2YAlC_2$ **n)** $Nb_2ZrAlC_2$ **o)** $W_2HfAlC_2$ **p)** $W_2NbAlC_2$ **q)** $W_2ScAlC_2$ **r)** $W_2TaAlC_2$ **s)** $W_2TiAlC_2$ **t)** $W_2VAlC_2$ **u)** $W_2YAlC_2$ **v)** $W_2ZrAlC_2$.

We conducted a series of ab initio molecular dynamics (AIMD) simulations, with the results presented in Figures 6S and 7S below. The calculations were carried out using a 2×2×1 supercell, and the structures were subjected to a temperature of 300 K for a duration of 5 picoseconds. Analysis of the simulation trajectories suggests the potential coexistence of an additional stable structure exhibiting Cc symmetry (group No. 9), which arises from a slight distortion of the hexagonal M' layer into parallel zigzag-like chains during the AIMD simulations. Notably, our simulations indicate that such a phase exists in the experimentally synthesized $Mo_2VAlC_2$. To assess the relative stability of the initial ($P6_3/mmc$) structure and the newly identified phase, we performed high-precision optimizations on both configurations. Following the optimization, we compared their total energies, revealing that the initial structure is approximately -0.005 eV/atom more stable than the newly discovered phase, as detailed in Table 4S. Such small energy difference suggests that the investigated o-MAX phases may possess two local minima that could be realized under specific experimental conditions.

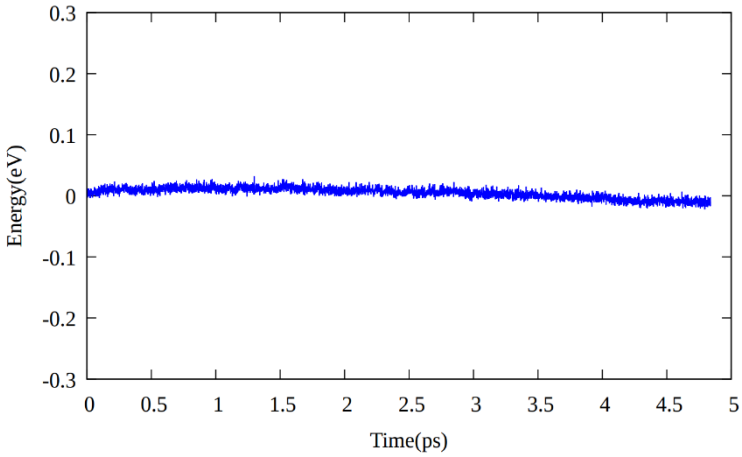
**Table4S.** Compare energy of compounds in the two competitive phases.

| **Material** | **Total energy (eV) per atom for new optimized structure $E_1$** | **Total energy (eV) per atom for original structure $E_2$** | **$E_2$-$E_1$ (eV/atom)** |
|---|---|---|---|
| **$W_2VAlC_2$** | -1114.2019348 | -1114.2092611 | -0.0073263 |
| **$W_2TaAlC_2$** | -1102.3201313 | -1102.3229161 | -0.0027848 |
| **$W_2NbAlC_2$** | -1051.8351656 | -1051.839192 | -0.0040264 |
| **$Mo_2NbAlC_2$** | -968.4465329 | -968.49461865 | -0.04808575 |

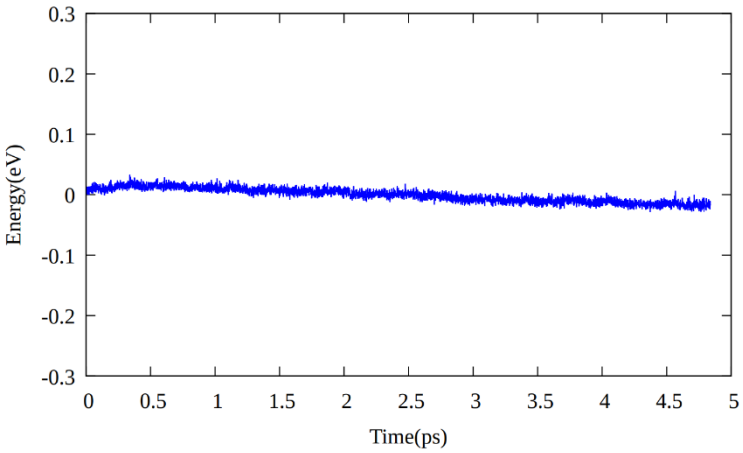
**a) $W_2VAlC_2$**



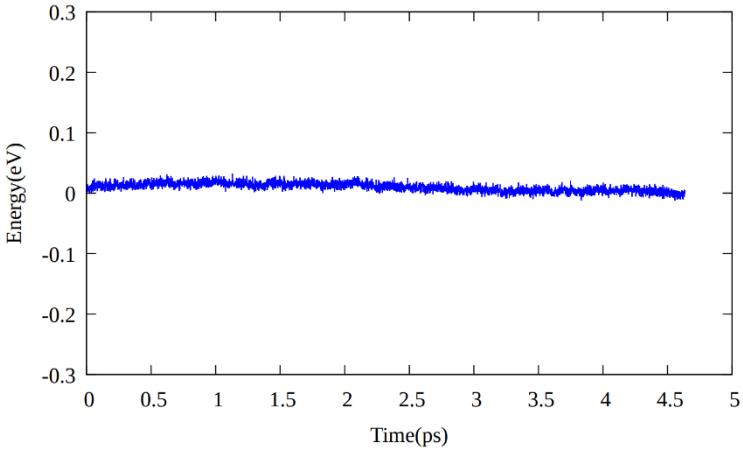

**b) $W_2TaAlC_2$**



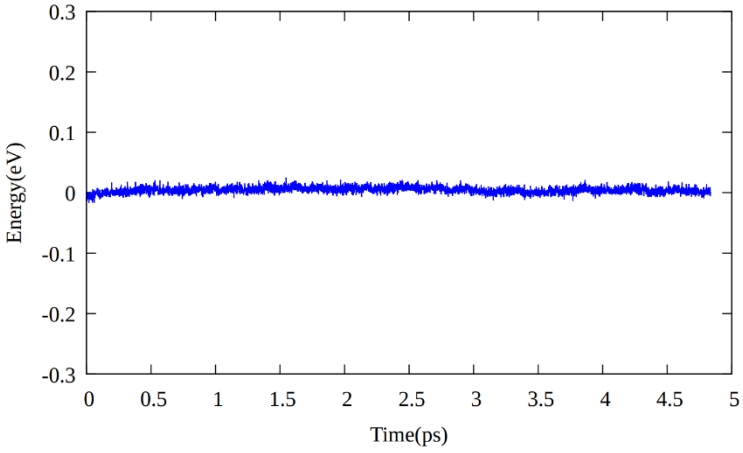

**c) $W_2NbAlC_2$**



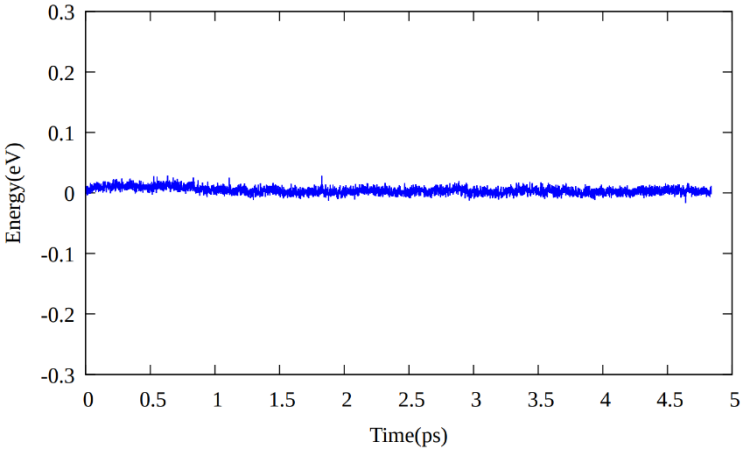

**d) $Mo_2NbAlC_2$**




**e) $Mo_2TaAlC_2$**




**f) $Mo_2TiAlC_2^*$**



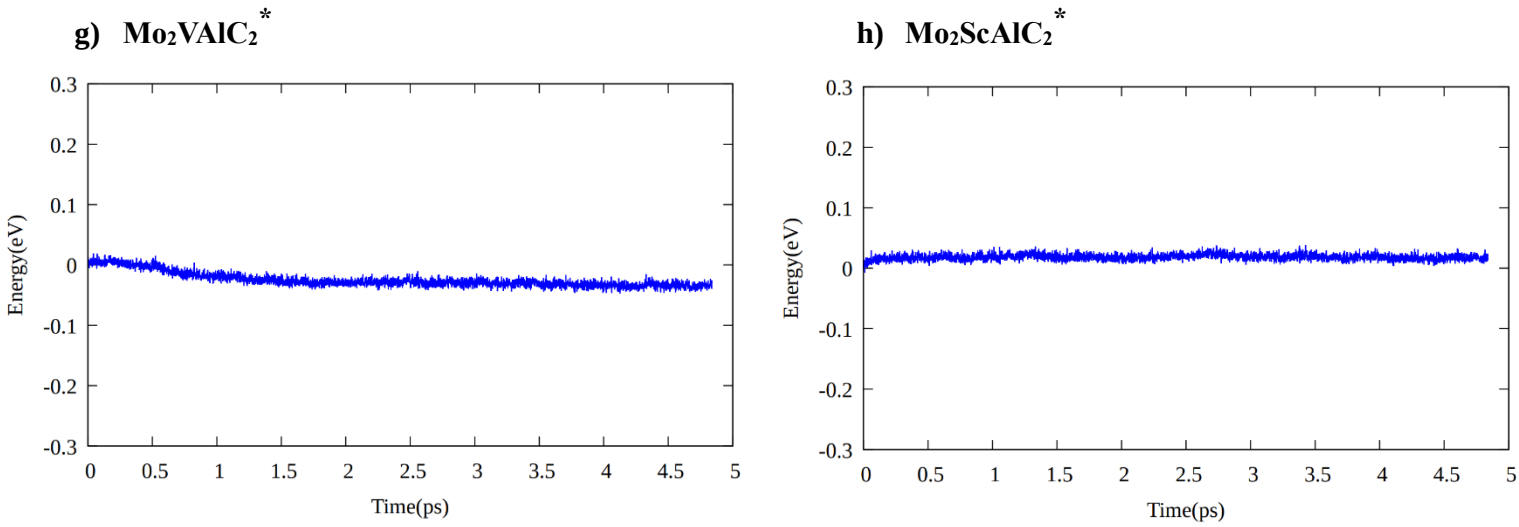



**Fig 6S.** ab initio molecular dynamics simulations for 6 of high-$T_c$ compounds. According to Table 4S after optimization the original structure is more stable. It is worth noting that $Mo_2ScAlC_2$, $Mo_2VAlC_2$, and $Mo_2TiAlC_2$ have been experimentally synthesized.

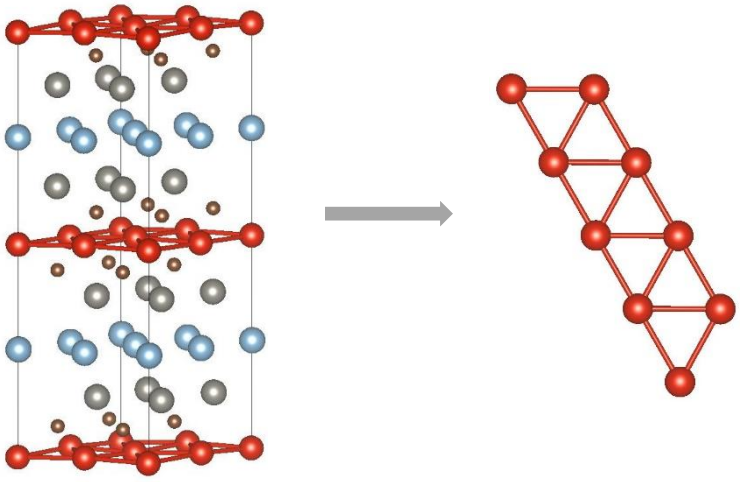


**Fig 7S**. The relaxed structure after Molecular Dynamic (MD) simulation.

The POSCAR file of the aforementioned competitive phase is placed below.

POSCAR File – o-MAX phase

1.0

| | | |
|---|---|---|
| 5.7901010079 | 0.0450632755 | -0.0042953188 |
| -2.8556289743 | 4.9470496107 | 0.0081578174 |
| -0.0104038889 | 0.0176582096 | 19.6674049400 |

| W | V | Al | C |
|---|---|---|---|
| 16 | 8 | 8 | 16 |

Direct

| | | |
|---|---|---|
| 0.150363609 | 0.323806494 | 0.361136973 |
| 0.154054075 | 0.823943794 | 0.369243205 |
| 0.650656939 | 0.328727841 | 0.364083648 |
| 0.651909113 | 0.828518093 | 0.363982737 |
| 0.347618669 | 0.176387906 | 0.864010453 |
| 0.348928779 | 0.677950621 | 0.864064932 |
| 0.849147618 | 0.173467040 | 0.861286342 |
| 0.845439434 | 0.669982851 | 0.869399548 |
| 0.347196609 | 0.172950357 | 0.636771679 |
| 0.351673067 | 0.681993127 | 0.635866582 |
| 0.847747564 | 0.176422745 | 0.639736772 |
| 0.851625502 | 0.682630718 | 0.633023143 |
| 0.151413202 | 0.328077972 | 0.139731422 |
| 0.147619247 | 0.830406368 | 0.133084401 |
| 0.647478461 | 0.329835981 | 0.135714456 |
| 0.652064323 | 0.825211465 | 0.136716112 |
| 0.011261389 | 0.003303064 | 0.980056047 |
| 0.997071147 | 0.489069551 | 0.017183051 |
| 0.496078610 | 0.001043542 | 0.019759573 |
| 0.515027463 | 0.502324700 | 0.979226410 |
| 0.988500297 | 0.992294371 | 0.479914129 |
| 0.001970237 | 0.491599530 | 0.517178178 |
| 0.484129012 | 0.986583471 | 0.479147017 |
| 0.503082812 | 0.504609346 | 0.519764781 |
| 0.988568783 | 0.982471228 | 0.251127779 |
| 0.986512959 | 0.507055581 | 0.250107944 |
| 0.477116823 | 0.991637170 | 0.249749944 |
| 0.494173586 | 0.498828769 | 0.250491142 |
| 0.010436894 | 0.993258297 | 0.751190186 |
| 0.012243116 | 0.520262599 | 0.750201821 |
| 0.505490899 | 0.004775260 | 0.750557184 |
| 0.522137046 | 0.514320433 | 0.749840975 |
| 0.161080286 | 0.352150291 | 0.932917833 |
| 0.184903264 | 0.837583363 | 0.937945485 |
| 0.673255920 | 0.307906836 | 0.938813031 |
| 0.648449361 | 0.876650214 | 0.939358175 |
| 0.351581901 | 0.229029909 | 0.439405292 |
| 0.326119483 | 0.633887589 | 0.438704014 |
| 0.838768542 | 0.191197962 | 0.432922453 |
| 0.815277815 | 0.653039813 | 0.437865764 |
| 0.320860237 | 0.180276200 | 0.062779106 |
| 0.339245319 | 0.644096911 | 0.065557316 |
| 0.839184999 | 0.153391749 | 0.067892849 |
| 0.819993079 | 0.663176119 | 0.060692627 |
| 0.160172433 | 0.313972533 | 0.567887664 |
| 0.179565951 | 0.842563331 | 0.560776114 |
| 0.659708500 | 0.304140955 | 0.565505147 |
| 0.678242743 | 0.859116971 | 0.562803328 |

To illustrate the feasibility of the experimental synthesis of o-MAX phases, we have conducted a comparison of the formation energies of our high $T_c$ o-MAX phases with their potential competitive binary phases, as detailed below. We have also done the same analysis for the experimentally synthesized structures. The results are summarized in Table 4S.

$$\Delta \mathbf{H_{W_2VAlC_2}}$$

$$\Delta \mathbf{H_1} = E_{W_2VAlC_2} - \left(\frac{1}{11}E_{VAl_3} + \frac{5}{11}E_{V_2C} + E_{W_2C} + \frac{2}{11}E_{Al_4C_3}\right)$$

$$\Delta \mathbf{H_2} = E_{W_2VAlC_2} - \left(\frac{1}{17}E_{WAl_3} + \frac{1}{2}E_{V_2C} + \frac{33}{34}E_{W_2C} + \frac{3}{17}E_{Al_4C_3}\right)$$

$$\Delta \mathbf{H_3} = E_{W_2VAlC_2} - \left(\frac{7}{37}E_{VAl_3} + \frac{5}{37}E_{V_6C_5} + E_{W_2C} + \frac{4}{37}E_{Al_4C_3}\right)$$

$$\Delta \mathbf{H_4} = E_{W_2VAlC_2} - \left(\frac{2}{13}E_{WAl_5} + \frac{2}{13}E_{V_2C} + \frac{24}{13}E_{WC} + \frac{3}{13}E_{V_3Al}\right)$$

$$\Delta \mathbf{H_5} = E_{W_2VAlC_2} - \left(\frac{7}{51}E_{WAl_5} + \frac{17}{102}E_{V_6C_5} + \frac{95}{102}E_{W_2C} + \frac{4}{51}E_{Al_4C_3}\right)$$

$$\Delta \mathbf{H_6} = E_{W_2VAlC_2} - \left(\frac{10}{69}E_{WAl_5} + \frac{2}{69}E_{V_6C_5} + \frac{128}{69}E_{WC} + \frac{19}{69}E_{V_3Al}\right)$$

$$\Delta \mathbf{H_{W_2TaAlC_2}}$$

$$\Delta \mathbf{H_1} = E_{W_2TaAlC_2} - \left(\frac{1}{7}E_{Ta_2Al} + \frac{5}{14}E_{Ta_2C} + E_{W_2C} + \frac{3}{14}E_{Al_4C_3}\right)$$

$$\Delta \mathbf{H_2} = E_{W_2TaAlC_2} - \left(\frac{1}{8}E_{Ta_2C} + \frac{3}{8}E_{Ta_2Al} + \frac{15}{8}E_{WC} + \frac{1}{8}E_{WAl_5}\right)$$

$$\Delta \mathbf{H_3} = E_{W_2TaAlC_2} - \left(\frac{5}{11}E_{TaC} + \frac{3}{11}E_{Ta_2Al} + E_{W_2C} + \frac{2}{11}E_{Al_4C_3}\right)$$

$$\Delta \mathbf{H_4} = E_{W_2TaAlC_2} - \left(\frac{10}{13}E_{TaC} + \frac{3}{13}E_{TaAl_3} + E_{W_2C} + \frac{1}{13}E_{Al_4C_3}\right)$$

$$\Delta \mathbf{H_5} = E_{W_2TaAlC_2} - \left(\frac{1}{2}E_{Ta_2C} + \frac{1}{17}E_{WAl_5} + \frac{33}{34}E_{W_2C} + \frac{3}{17}E_{Al_4C_3}\right)$$

$$\Delta \mathbf{H_6} = E_{W_2TaAlC_2} - \left(E_{TaC} + \frac{3}{17}E_{WAl_5} + \frac{31}{34}E_{W_2C} + \frac{1}{34}E_{Al_4C_3}\right)$$

$$\Delta \mathbf{H_{W_2NbAlC_2}}$$

$$\Delta \mathbf{H_1} = E_{W_2NbAlC_2} - \left(\frac{1}{7}E_{Nb_2Al} + \frac{5}{14}E_{Nb_2C} + E_{W_2C} + \frac{3}{14}E_{Al_4C_3}\right)$$

$$\Delta \mathbf{H_2} = E_{W_2NbAlC_2} - \left(\frac{1}{2}E_{Nb_2C} + \frac{1}{17}E_{WAl_5} + \frac{33}{34}E_{W_2C} + \frac{3}{17}E_{Al_4C_3}\right)$$

$$\Delta \mathbf{H_3} = E_{W_2NbAlC_2} - \left(\frac{5}{11}E_{Nb_2C} + \frac{1}{11}E_{NbAl_3} + E_{W_2C} + \frac{2}{11}E_{Al_4C_3}\right)$$

$$\Delta \mathbf{H_4} = E_{W_2NbAlC_2} - \left(\frac{5}{37}E_{Nb_6C_5} + \frac{7}{37}E_{NbAl_3} + E_{W_2C} + \frac{4}{37}E_{Al_4C_3}\right)$$

$$\Delta \mathbf{H_5} = E_{W_2NbAlC_2} - \left(\frac{1}{44}E_{Nb_6C_5} + \frac{19}{44}E_{Nb_2Al} + \frac{5}{44}E_{WAl_5} + \frac{83}{44}E_{WC}\right)$$

$$\Delta \mathbf{H_6} = E_{W_2NbAlC_2} - \left(\frac{1}{3}E_{Nb_2C} + \frac{1}{3}E_{W_2C} + \frac{1}{3}E_{NbAl_3} + \frac{4}{3}E_{WC}\right)$$

$$\Delta H_{Mo_2VAlC_2}$$

$$\Delta H_1 = E_{Mo_2VAlC_2} - \left(\frac{1}{11}E_{VAl_3} + \frac{5}{11}E_{V_2C} + E_{Mo_2C} + \frac{2}{11}E_{Al_4C_3}\right)$$

$$\Delta H_2 = E_{Mo_2VAlC_2} - \left(\frac{1}{13}E_{V_6C_5} + \frac{7}{39}E_{V_3Al} + E_{Mo_2C} + \frac{8}{39}E_{Al_4C_3}\right)$$

$$\Delta H_3 = E_{Mo_2VAlC_2} - \left(\frac{5}{37}E_{V_6C_5} + \frac{5}{11}E_{VAl_3} + E_{Mo_2C} + \frac{4}{37}E_{Al_4C_3}\right)$$

$$\Delta H_4 = E_{Mo_2VAlC_2} - \left(\frac{1}{9}E_{V_3Al} + \frac{1}{3}E_{V_2C} + E_{Mo_2C} + \frac{2}{9}E_{Al_4C_3}\right)$$

$$\Delta H_5 = E_{Mo_2VAlC_2} - \left(\frac{1}{2}E_{V_2C} + \frac{1}{9}E_{Mo_3Al} + \frac{15}{18}E_{Mo_2C} + \frac{2}{9}E_{Al_4C_3}\right)$$

$$\Delta H_6 = E_{Mo_2VAlC_2} - \left(\frac{1}{6}E_{V_6C_5} + \frac{7}{27}E_{Mo_3Al} + \frac{33}{54}E_{Mo_2C} + \frac{5}{27}E_{Al_4C_3}\right)$$

$$\Delta H_{Mo_2NbAlC_2}$$

$$\Delta H_1 = E_{Mo_2NbAlC_2} - \left(\frac{1}{7}E_{Nb_2Al} + \frac{5}{14}E_{Nb_2C} + E_{Mo_2C} + \frac{3}{14}E_{Al_4C_3}\right)$$

$$\Delta H_2 = E_{Mo_2NbAlC_2} - \left(\frac{5}{58}E_{Nb_6C_5} + \frac{14}{58}E_{Nb_2Al} + E_{Mo_2C} + \frac{11}{58}E_{Al_4C_3}\right)$$

$$\Delta H_3 = E_{Mo_2NbAlC_2} - \left(\frac{1}{20}E_{Nb_6C_5} + \frac{7}{10}E_{Nb} + E_{Mo_2C} + \frac{1}{4}E_{Al_4C_3}\right)$$

$$\Delta H_4 = E_{Mo_2NbAlC_2} - \left(\frac{1}{6}E_{Nb_6C_5} + \frac{33}{54}E_{Mo2C} + \frac{7}{27}E_{Mo_3Al} + \frac{5}{27}E_{Al_4C_3}\right)$$

$$\Delta H_5 = E_{Mo_2NbAlC_2} - \left(\frac{1}{2}E_{Nb_2C} + \frac{15}{18}E_{Mo_2C} + \frac{1}{9}E_{Mo_3Al} + \frac{2}{9}E_{Al_4C_3}\right)$$

$$\Delta H_6 = E_{Mo_2NbAlC_2} - \left(\frac{5}{11}E_{Nb_2C} + \frac{1}{11}E_{NbAl_3} + E_{Mo_2C} + \frac{2}{11}E_{Al_4C_3}\right)$$

$$\Delta H_{Mo_2ScAlC_2}$$

$$\Delta H_1 = E_{Mo_2ScAlC_2} - \left(\frac{1}{7}E_{Sc_2Al} + \frac{5}{14}E_{Sc_2C} + E_{Mo_2C} + \frac{3}{14}E_{Al_4C_3}\right)$$

$$\Delta H_2 = E_{Mo_2ScAlC_2} - \left(\frac{1}{2}E_{Sc_2C} + \frac{2}{9}E_{Mo_3Al} + \frac{15}{18}E_{Mo_2C} + \frac{2}{9}E_{Al_4C_3}\right)$$

$$\Delta H_3 = E_{Mo_2ScAlC_2} - \left(\frac{1}{4}E_{Sc_4C_3} + \frac{2}{9}E_{Mo_3Al} + \frac{2}{3}E_{Mo_2C} + \frac{7}{36}E_{Al_4C_3}\right)$$

$$\Delta H_4 = E_{Mo_2ScAlC_2} - \left(\frac{1}{6}E_{Sc_4C_3} + E_{Mo_2C} + \frac{1}{3}E_{ScAl} + \frac{1}{6}E_{Al_4C_3}\right)$$

$$\Delta H_5 = E_{Mo_2ScAlC_2} - \left(\frac{7}{36}E_{Sc_4C_3} + E_{Mo_2C} + \frac{2}{9}E_{ScAl_2} + \frac{5}{36}E_{Al_4C_3}\right)$$

$$\Delta H_6 = E_{Mo_2ScAlC_2} - \left(\frac{5}{24}E_{Sc_4C_3} + E_{Mo_2C} + \frac{1}{6}E_{ScAl_3} + \frac{1}{8}E_{Al_4C_3}\right)$$

$$\Delta H_{Mo_2TiAlC_2}$$

$$\Delta H_1 = E_{Mo_2TiAlC_2} - \left(\frac{1}{8}E_{TiAl_2} + \frac{7}{16}E_{Ti_2C} + E_{Mo_2C} + \frac{3}{16}E_{Al_4C_3}\right)$$

$$\Delta H_2 = E_{Mo_2TiAlC_2} - \left(\frac{5}{46}E_{Ti_8C_5} + \frac{3}{23}E_{TiAl_3} + E_{Mo_2C} + \frac{7}{46}E_{Al_4C_3}\right)$$

$$\Delta H_3 = E_{Mo_2TiAlC_2} - \left(\frac{1}{2}E_{Ti_2C} + \frac{1}{9}E_{Mo_3Al} + \frac{15}{18}E_{Mo_2C} + \frac{2}{9}E_{Al_4C_3}\right)$$

$$\Delta H_4 = E_{Mo_2TiAlC_2} - \left(E_{TiC} + \frac{1}{3}E_{Mo_3Al} + \frac{1}{2}E_{Mo_2C} + \frac{1}{6}E_{Al_4C_3}\right)$$

$$\Delta\mathbf{H_5} = E_{Mo_2TiAlC_2} - \left(\frac{10}{13}E_{TiC} + \frac{3}{13}E_{TiAl_3} + E_{Mo_2C} + \frac{1}{13}E_{Al_4C_3}\right)$$

$$\Delta\mathbf{H_6} = E_{Mo_2TiAlC_2} - \left(\frac{4}{7}E_{TiC} + \frac{3}{7}E_{TiAl} + E_{Mo_2C} + \frac{1}{7}E_{Al_4C_3}\right)$$

These results in Table 5S indicate that the calculated relative formation energies of these compounds are either negative or very small positive. According to the recent computational screening on the inorganic crystal structure database (ICSD), 80% of experimentally synthesized compounds possess instability of ΔH less than 0.036 eV per atom [G. Bergerhoff, R. Hundt and R. Sievers, J. Chem. Inf. Comput. Sci., 1983, 23, 66–69]. Assuming this value as a stability criterion (i.e., $\Delta H < 0.036$ eV per atom), it is expected that o-MAX phases with high $T_c$ have possibility for the experimental synthesis. We have added the above discussion to the manuscript and the Table to the Supplementary information.

**Table 5S.** The relative formation energies of the o-MAX phases have been calculated and compared to their corresponding binary phases. The binary phases were sourced from the database of the Materials Project. The asterisk (*) denotes that the o-MAX phases were synthesized through experimental methods.

| Materials | Available experimental phases | ΔH (eV/atom) |
|---|---|---|
| $W_2VAlC_2$ | $VAl_3$, $V_2C$, $W_2C$, $Al_4C_3$, $V_6C_5$, $V_3Al$, WC, $WAl_5$, $V_6C_5$ | $\Delta H_1$ = -0.01 $\Delta H_2$ = -0.015 $\Delta H_3$= 0.022 $\Delta H_4$ = -0.009 $\Delta H_5$ = 0.036 $\Delta H_6$ = -0.013 |
| $W_2TaAlC_2$ | $Ta_2Al$, $Ta_2C$, TaC, $W_2C$, $Al_4C_3$, WC, $WAl_5$ | $\Delta H_1$ = -0.043 $\Delta H_2$ =-0.033 $\Delta H_3$ = -0.062 $\Delta H_4$ = 0.007 $\Delta H_5$ = -0.023 $\Delta H_6$ =0.030 |
| $W_2NbAlC_2$ | $Nb_2Al$, $Nb_2C$, $W_2C$, $Al_4C_3$, WC, $WAl_5$, $Nb_6C_5$ | $\Delta H_1$ = -0.04 $\Delta H_2$ =-0.033 $\Delta H_3$ = -0.027 $\Delta H_4$ = 0.029 $\Delta H_5$ = -0.008 $\Delta H_6$ =0.042 |
| $Mo_2VAlC_2$ | $VAl_3$, $V_2C$, $Mo_2C$, $Al_4C_3$, $V_6C_5$, $V_3Al$, $Mo_3Al$, $V_6C_5$ | $\Delta H_1$ = -0.058 $\Delta H_2$ = -0.102 $\Delta H_3$ =-0.034 $\Delta H_4$ = -0.105 $\Delta H_5$ = -0.070 $\Delta H_6$ =-0.016 |
| *$Mo_2NbAlC_2$ | $Nb_2Al$, $Nb_2C$, $Mo_2C$, $Al_4C_3$, $Mo_3Al$, $Nb_6C_5$ | $\Delta H_1$ = -0.110 $\Delta H_2$ =-0.091 $\Delta H_3$ =-0.162 $\Delta H_4$ =-0.022 $\Delta H_5$ = -0.087 $\Delta H_6$ =-0.083 |
| *$Mo_2ScAlC_2$ | $Sc_2Al$, $Sc_2C$, $Mo_2C$, $Al_4C_3$, $Sc_4C_3$, $Mo_3Al$, ScAl, $ScAl_2$, $ScAl_3$ | $\Delta H_1$ = -0.194 $\Delta H_2$ =-0.181 $\Delta H_3$ =-0.142 $\Delta H_4$ = -0.156 $\Delta H_5$ =-0.139 $\Delta H_6$ =-0.135 |
| *$Mo_2TiAlC_2$ | $TiAl_2$, $Ti_2C$, $Mo_2C$, $Al_4C_3$, TiC, TiAl, $TiAl_3$, $Ti_8C_5$, $Mo_3Al$ | $\Delta H_1$ = -0.245 $\Delta H_2$ = -0.174 $\Delta H_3$ = -0.181 $\Delta H_4$ =-0.211 $\Delta H_5$ =-0.095 $\Delta H_6$ =-0.124 |