Electronic and vibrational properties of V$_2$C-based MXene: from experiments to first-principles modelling

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MXenes are new kind of 2D materials produced by selectively etching layers of sp-elements from their corresponding 3D MAX phase (Fig.1.a). Most MXenes exhibit a unique combination of excellent physical and chemical properties [1] which make them very promising 2D materials for energy devices [2] such as hydrogen storage, Li-ion batteries [3], and electrochemical capacitors [4].

In the present work, the electronic and vibrational properties of pristine bare V$_2$C (Fig.1.b-c) and terminated V$_2$CT$_2$ (T=F, O, OH) (Fig.1.d) 2D layers are investigated based on Density Functional Theory approach. Firstly, the atomic structures of V$_2$C-based MXene phases are optimized (trigonal P-3m1 space group). Secondly, electronic band structures are computed indicating that V$_2$C is metallic phase as well as V$_2$CF$_2$, V$_2$CO$_2$, and V$_2$C(OH)$_2$. Thirdly, phonons, which play a major role in thermal and electronic conductivity of MXenes [5], are computed thanks to Density Functional Perturbation Theory. Both Raman (E$_g$, A$_{1g}$) and infrared active (E$_u$, A$_{2u}$) vibrational modes are predicted ab initio and are found to be in good agreement (maximal deviation 10%) with experimental Raman spectra (Fig.2). However, it is still challenging to exfoliate pure V$_2$C MXene monosheets from the 3D MAX phase. Indeed, chemically exfoliated MXenes usually contain heterogeneous terminal groups [5]. The small discrepancy between experimental and simulated phonon frequencies reported here could be explained by the presence of a mixture of the four types of simulated structures in the exfoliated sample.

References

Figures

**Figure 1:** SEM images of (a) MAX V$_2$AlC, (b) MXene V$_2$C, and ab initio atomic structures of (c) V$_2$C and (d) terminated V$_2$CT$_2$ (T=F, O, OH).

**Figure 2:** Experimental Raman spectra of the MAX V$_2$AlC (blue) and the corresponding exfoliated MXene V$_2$C (red) phase. Ab initio Raman frequencies of pure V$_2$C (green) are also included for comparison.