Electronic and vibrational properties of V_2C-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_2C (Fig.1.b-c) and terminated V_2CT_2 (T=F,O,OH) (Fig.1.d) 2D layers are investigated based on Density Functional Theory approach. Firstly, the atomic structures of V_2C-based MXene phases are optimized (trigonal P-3m1 space group). Secondly, electronic band structures are computed indicating that V_2C is metallic phase as well as V_2CF_2, V_2CO_2, and $V_2C(OH)_2$. 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_2C 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

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Figures

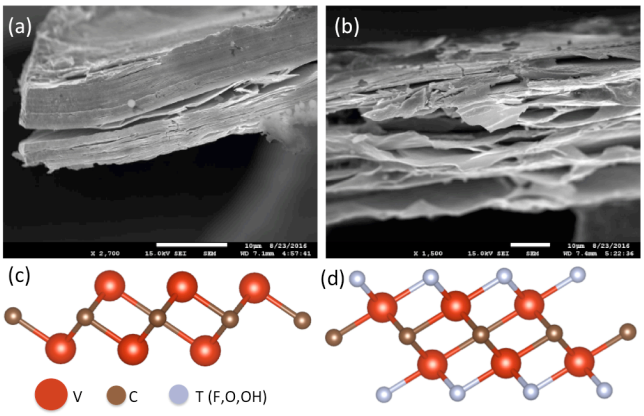

Figure 1: SEM images of (a) MAX V_2AlC, (b) MXene V_2C, and *ab initio* atomic structures of (c) V_2C and (d) terminated V_2CT_2 (T=F,O,OH).

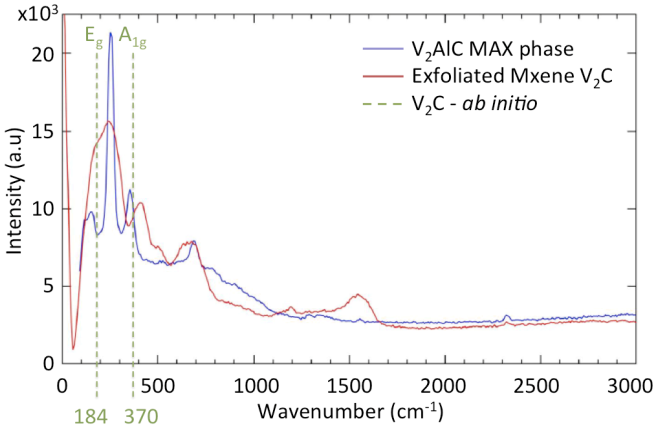

Figure 2: Experimental Raman spectra of the MAX V_2AlC (blue) and the corresponding exfoliated MXene V_2C (red) phase. *Ab initio* Raman frequencies of pure V_2C (green) are also included for comparison.