Machine-Learned Interatomic Potentials for Transition Metal Dichalcogenide $Mo_{1-x}W_xS_{2-2y}Se_{2y}$ Alloys

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Machine Learned Interatomic Potentials (MLIP) harness the accuracy of Density Functional Theory (DFT) in conjunction with the computational efficiency and scalability of interatomic potentials. This synergy enables theoretical spectroscopy to delve into larger and more intricate systems than previously feasible with DFT alone. In this work, we train an MLIP for quaternary Transition Metal Dichalcogenide (TMDC) alloy systems of the form $Mo_{1-x}W_xS_{2-2y}Se_{2y}$, using the equivariant Neural Network (NN) MACE[1]. We demonstrate the ability of this potential to calculate vibrational properties of alloy TMDCs including phonon spectra for pure monolayers, and VDOS and Raman spectra for alloys, retaining DFT- level accuracy while greatly extending feasible system size and degree of sampling over alloy configurations. We are able to characterise the Raman active modes across the whole range of concentration, particularly for the "disorder induced" modes. This potential can serve as a tool to aid experimentalists in studying and designing TMDC alloys for future applications.

References

[1] I. Batatia, D. P. Kovacs, G. N. C. Simm, C. Ortner, and G. Csanyi, MACE: Higher order equivariant message passing neural networks for fast and accurate force fields, Advances in Neural Information Processing Systems (2022)
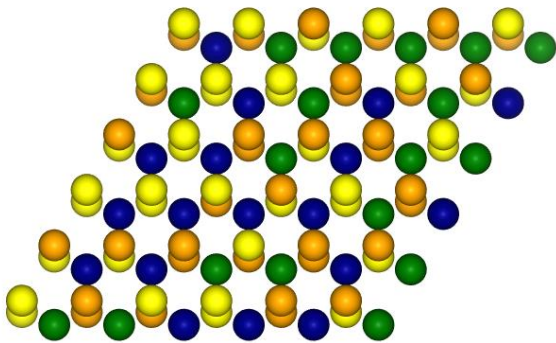
Figures

Figure 1. Top view of a random 6x6 $Mo_{0.5}W_{0.5}SSe$ alloy supercell. Mo, W, S and Se atoms are denoted by green, blue, yellow and orange colored balls.
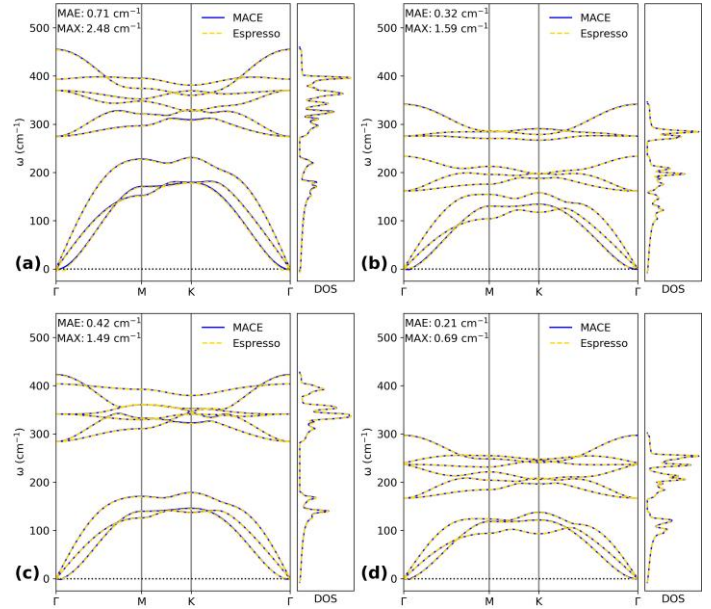

Figure 2. Phonon Dispersion curves and density of states for pure (a) MoS_2, (b) $MoSe_2$, (c) WS_2 and (d) WSe_2 monolayers calculated using DFT (dashed yellow lines) and MACE (solid blue lines).
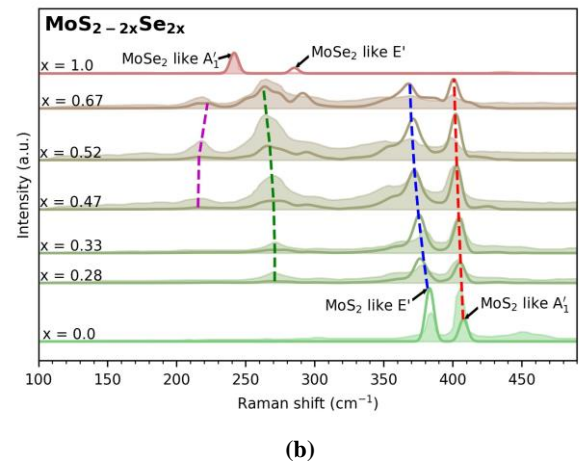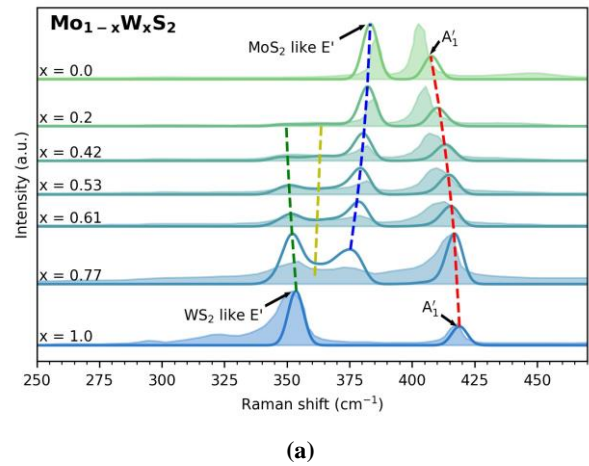

(a)

(b)

Figure 3. (a) Experimental (filled) and MACE calculated (line) Raman spectra of Mo1-xWS2 monolayers with different W compositions x **(b)** Experimental (filled) and MACE calculated (line) Raman spectra of WS2-2xSe2x monolayers with different Se compositions x. The colored dashed lines in (a) and (b) show frequency shift of different peaks.