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Phonon anharmonicity in supported single- and multi-layered WS_2 nanosheets – first principles and Raman investigation

The purpose of this work is the analysis of phonon anharmonicity in single- and multi-layered 2H-WS_2 nanosheets – a representative example among widely studied 2D transition metal dichalcogenides [1]. Anharmonicity has a significant impact on the temperature-dependent phonon properties: energies and lifetimes. For the supported samples, the substrate can further affect their vibrational properties. Although individual Raman studies with respect to temperature [2] and impact of the substrate – strain and charge doping [3] – are available for the most popular 2D materials (e.g., MoS_2), also for multi-layered nanosheets, a comprehensive ab-initio study of supported films with different numbers of layers is missing in the literature. Here, we present DFT simulations of temperature-dependent (0–500 K) phonon properties in 1–5 layered WS_2 nanosheets, including three-phonon interaction processes, thermal expansion, and substrate effects. We compare the simulated frequencies and bandwidths of the representative zone-center A_{1g} and $E_{2g}{}^1$ phonon modes to Raman spectra (75–500 K) measured in CVD-grown WS_2 thin films. We obtain a good agreement between ab-initio and experimental temperature trends (with respect to temperature-independent additive constants) in the case of A_{1g} (Figure 1) and high-temperature $E_{2g}{}^1$ phonons, whereas the low-temperature discrepancies for the $E_{2g}{}^1$ mode can be explained in terms of vertical strain distribution in multi-layered WS_2. Moreover, we simulate frequency shifts caused by interaction with the substrate, including mixed effects of charge doping and induced strain. The presented study of phonon anharmonicity is important due to its significant impact on thermal and electrical transport in nanosheets, including their potential applications. The results are also helpful for Raman characterization of supported WS_2 thin films at different temperatures.

References

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[2] S. Zhu & W. Zheng, The Journal of Physical Chemistry Letters, **12** (2021) 5261.
[3] W. H. Chae, J. D. Cain, E. D. Hanson, A. A. Murthy, & V. P. Dravid, Applied Physics Letters, **111** (2017) 143106.

Figures

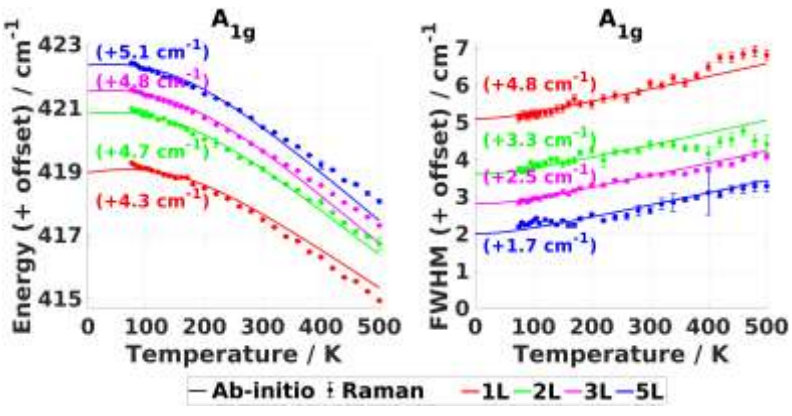

Figure 1: Temperature-dependent ab-initio and Raman results of energy and bandwidth of zone-center A_{1g} phonon mode in 1–5 layered WS_2 films. For clarity, the simulation results are shifted by additive constants (shown in brackets).