First-principles calculation of Raman spectra of 733 different 2D materials

A. Taghizadeh
U. Leffers, T.G. Pedersen, K. S. Thygesen
Department of Materials and Production, Aalborg University, Aalborg East, Denmark
CAMD, Department of Physics, Technical University of Denmark (DTU), Kgs. Lyngby, Denmark
ata@nano.aau.dk

Raman spectroscopy is a pivotal experimental technique for two-dimensional (2D) materials including identification of material composition and phase as well as probing various properties such as thickness (number of layers), interlayer coupling, and sample quality [1]. From a theoretical point of view, *ab initio* methods such as density-functional theory (DFT) have been successfully employed and typically yield results in good agreement with experiments. In the realm of 2D materials, *ab initio* Raman calculations have only been done for a handful of popular 2D materials such as graphene, MoS_2, and WS_2. We present our results on first-principles calculations of Raman spectra of 733 monolayers [2] selected as the most stable 2D crystals from the computational 2D materials database (C2DB). The calculations are based on our efficient DFT implementation using a localized atomic orbital (LCAO) basis set in combination with third-order perturbation theory. All computed Raman spectra (at three excitation wavelengths and nine possible polarization setups) will be available through the C2DB website. We benchmark the computational scheme against available experimental data and present several examples. Using the library of Raman spectra, we carefully test the inverse Raman problem, i.e. identifying the underlying material by comparing a given experimental spectrum to the library. For this purpose, we propose a simple but effective approach based on the first and second moments of the Raman spectrum and demonstrate the feasibility of this approach for the cases of MoS_2 (in H-phase) and WTe_2 (in T'-phase) as two examples. For instance, in Figure 1, we illustrate the scattering plot of the Raman fingerprints (first moments and normalized standard deviations of Raman spectra) for 733 monolayers, which will be discussed in detail in the presentation. We believe that our results would be extremely interesting for both experimentalists and theoreticians in the field of 2D materials.

References

[1] A. C. Ferrari and D. M. Basko, Nat. Nanotechnol. 8 (2013), 235-246.
[2] A. Taghizadeh, U. Leffers, T. G. Pedersen, and K. S. Thygesen, arXiv:2001.06313.

Figures

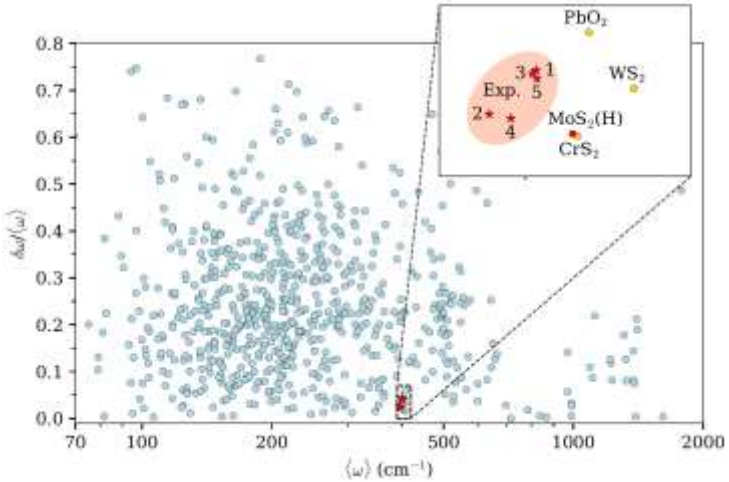

Figure 1: Scatter plot of the first Raman moment and normalized standard deviation for 733 calculated spectra (circles). For comparison, several experimental spectra for monolayer MoS_2 are shown (stars). Details will be discussed in the presentation.