Predicting Raman Spectra of Group III Nitride Superlattices with Density Functional Theory

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Measuring Vibrational Energy

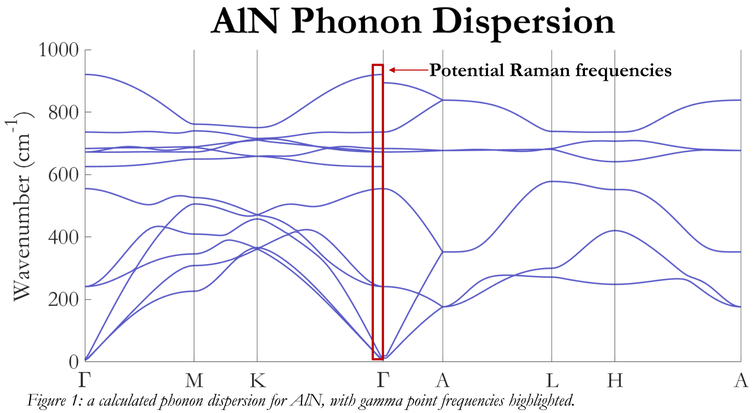


Figure 1: a calculated phonon dispersion for AlN, with gamma point frequencies highlighted.

A **phonon** describes the **transport of vibrational energy** in a material. A **phonon dispersion** depicts available frequencies in a system.

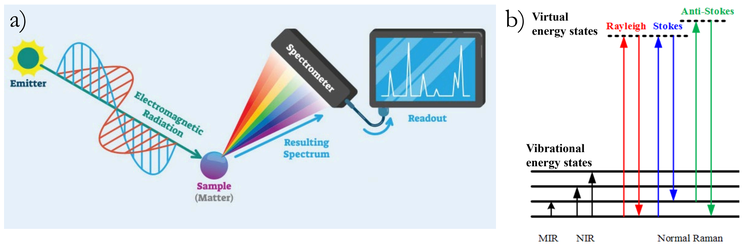


Figure 2: (a) diagram of Raman spectroscopy instrumentation [1] (b) depiction of energy states in Raman scattering [2]

Raman spectroscopy is a light-based, experimental method that can probe certain vibrational modes.

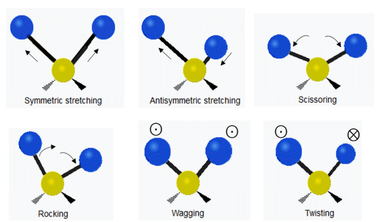


Figure 3: various vibrational motions [3]

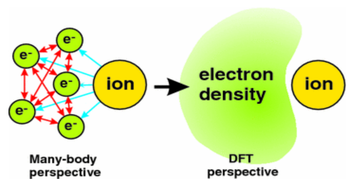
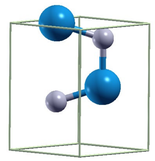


Figure 4: DFT electronic approximation [4]

Symmetry determines which frequencies are Raman active, due to changes in polarizability.

Density Functional Theory (DFT) can calculate phonon dispersions and Raman activity.

Raman Spectra of AlN & GaN



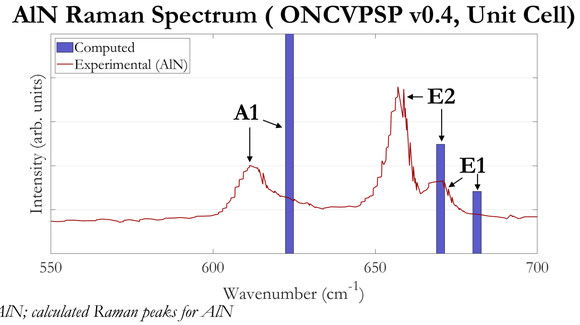
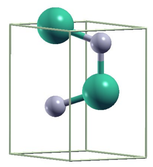


Figure 5: the unit cell for AlN; calculated Raman peaks for AlN

Simulation results for AlN place A1, E2, and E1 Raman peaks in the correct order, and locate these peaks with reasonable accuracy. Relative intensities, however, are not accurate.



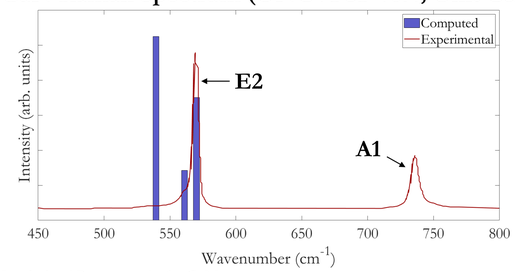
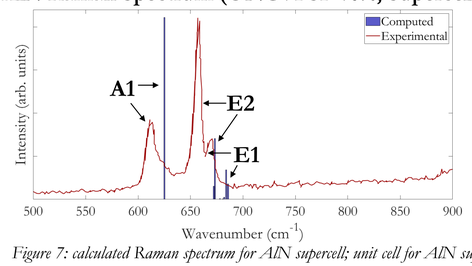


Figure 6: the unit cell for GaN; calculated Raman peaks for GaN

Calculation results for GaN correctly place the E2 Raman peak, but fail to capture the A1 peak at approximately 735 cm^{-1}.



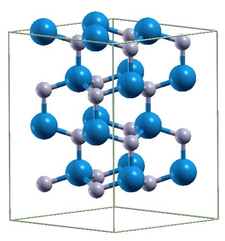
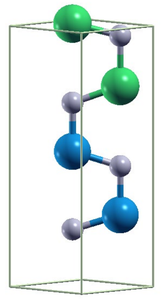


Figure 7: calculated Raman spectrum for AlN supercell; unit cell for AlN supercell

Simulating a larger system increases the number of calculated vibrational modes, which increases the detail of the spectra. Peak-like shapes begin to form, but concerns about relative intensities remain.

AlN-GaN Superlattice



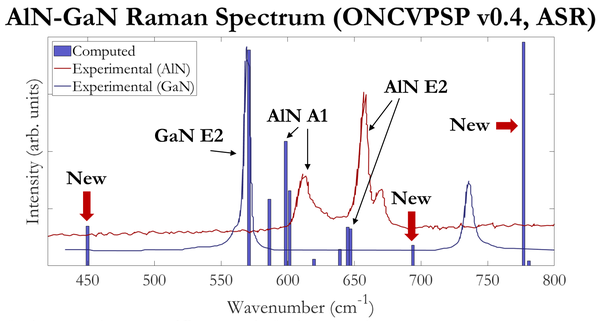


Figure 8: the unit cell for the superlattice; calculated Raman peaks for the superlattice

- DFT can accurately calculate **phonon frequencies** and **Raman-active modes.**
- Unique modes arise in the superlattice, representing potential **interface** or **hybrid modes**.

Frequency	Origin
450 cm^{-1}	New mode
571 cm^{-1}	GaN E2
601 cm^{-1}	AlN E1
647 cm^{-1}	AlN E2
694 cm^{-1}	New mode
781 cm^{-1}	New mode

Increasing the size of this system and improving convergence may yield improved intensity data and the calculation of additional active peaks, which would improve the trustworthiness of calculations.

Acknowledgments

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[1] Spectroscopy. (n.d.). PASCO Scientific. Retrieved August 2, 2021, from https://www.pasco.com/products/guides/what-is-spectroscopy
[2] Li, Zhiyun & Deen, M.J. & Selvaganapathy, Ponnambalam. (2014). Raman Spectroscopy for In-Line Water Quality Monitoring - Instrumentation and Potential. Sensors (Basel, Switzerland). 14. 17275-17303.10.3390/s140917275.
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[4] Lusk, Mark & Mattsson, Ann. (2011). High-performance computing for materials design to advance energy science. MRS Bulletin. 36. 10.1557/mrs.2011.30.