Polarised Raman and Second Harmonic Generation Microscopy for Probing Symmetry, Strain, and Layer Interactions in Two-Dimensional Materials

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Understanding the structural and electronic properties of two-dimensional (2D) materials requires characterisation techniques that are both non-destructive and highly sensitive to crystal symmetry, interlayer coupling, and local variations in strain or defects. In this work, we demonstrate how the combined use of polarised Raman spectroscopy and second harmonic generation (SHG) microscopy enables powerful multimodal analysis of 2D materials.

Polarised Raman spectroscopy provides orientation-dependent phonon responses that allow determination of crystal axes, identification of polytype variations, and mapping of strain or doping heterogeneity with sub-micron spatial resolution. Complementary SHG imaging offers symmetry-specific contrast that is highly sensitive to layer number, grain boundaries, and rotational domains, enabling rapid visualisation of crystal quality across large sample areas.

By correlating SHG and Raman signatures, we obtain insights that would be inaccessible to either technique alone. SHG maps guide the interpretation of polarisation-resolved Raman measurements, while Raman peak shifts and intensity variations reveal mechanical or electronic changes that are not SHG-active. Together, these modalities provide a coherent framework for quantitatively assessing growth quality, domain orientation, and local structural variations in emerging 2D heterostructures. This multimodal approach supports the development of reproducible, scalable characterisation workflows essential for integrating 2D materials into next-generation optoelectronic, quantum, and photonic devices.