IMAGING OF SEMICONDUCTORS USING SECOND HARMONIC GENERATION (SHG) MICROSCOPY

Combining the Chromacity 1040 femtosecond laser with an Edinburgh Instruments RMS1000 Confocal Microscope

Second Harmonic Generation

Second harmonic generation (SHG) microscopy is an imaging technique where the sample's ability to generate second harmonic light creates contrast in the image. SHG is a non-linear optical process where two photons with the same wavelength interact in a material to generate a photon with exactly half the wavelength of the incident photons. The generation of a SHG signal depends on the symmetry of the molecular structures in present a sample, with SHG only occurring when there is a noncentrosymmetry in the excitation focal volume. This sensitivity to symmetry makes SHG imaging a powerful technique in materials science where it can be used to image the grain boundaries and layer stacking in 2D semiconductors through the changes in crystal symmetry.

Experimental Configuration

SHG requires a very high excitation intensity which is achieved using mode-locked femtosecond pulsed lasers. Traditionally this has been titanium sapphire lasers, but femtosecond fiber lasers are becoming an increasingly attractive and economical alternative. The Edinburgh Instruments RMS1000 Confocal Raman Microscope has external laser coupling ports that enable the optical coupling of mode-locked femtosecond lasers into the microscope. An example experimental setup for SHG microscopy with the RMS1000 is shown below. The sample is excited by the femtosecond laser and the SHG signal acquired on the RMS1000 CCD camera.
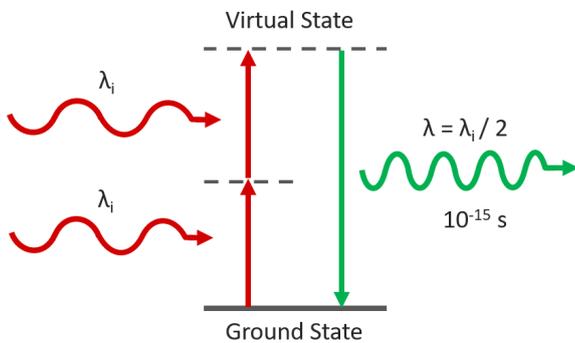

FIGURE 1. Schematic of the second harmonic process. This nonlinear optical process, causes two long wavelength photons to interact within a material to form new photons having twice the frequency of the input photons.
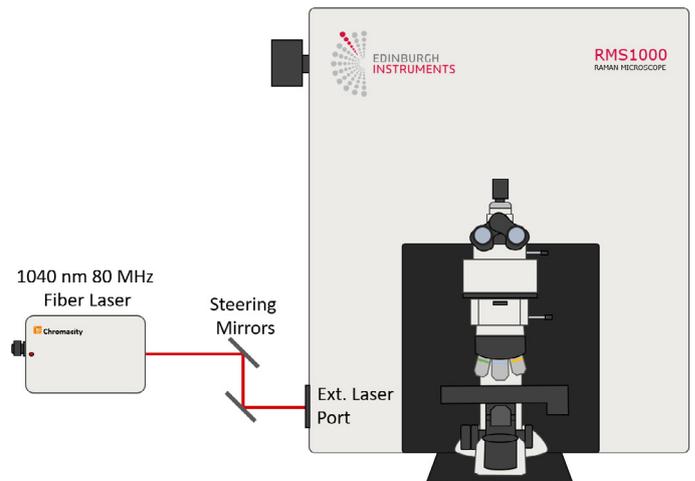

FIGURE 2. Example optical setup for SHG microscopy with the Edinburgh Instruments RMS1000. The laser excitation source is a Chromacity 1040 femtosecond fiber laser with an output wavelength of 1040 nm and a repetition rate of 80 MHz (Chromacity Ltd., UK).

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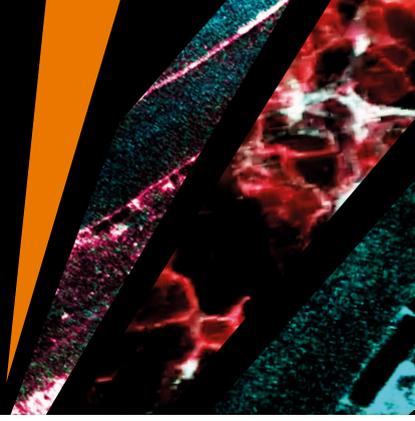
Examples of SHG Imaging with the RMS1000

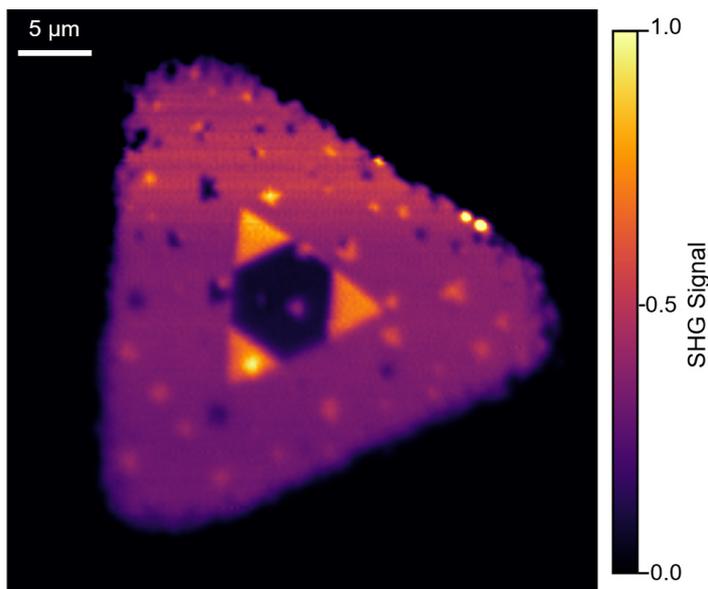### WSe_2 Flake

WS_2 Polycrystal

FIGURE 3. Second harmonic generation imaging of a WSe_2 flake grown using CVD. The SHG image reveals the presence of multiple layers in the centre of the flake. The main triangle is monolayer WSe_2 which has broken inversion symmetry (non-centrosymmetric) and therefore generates a SHG response. The three smaller bright triangles are bilayer WSe_2 where the second layer has the same crystal orientation as the monolayer and broken inversion symmetry is maintained so the SHG response of the layers is additive. The dark region in the centre is likely trilayer WSe_2 where the crystal growth is 180 degrees opposite to the monolayer and bilayer resulting in inversion symmetry and a significant decrease of the SHG signal.
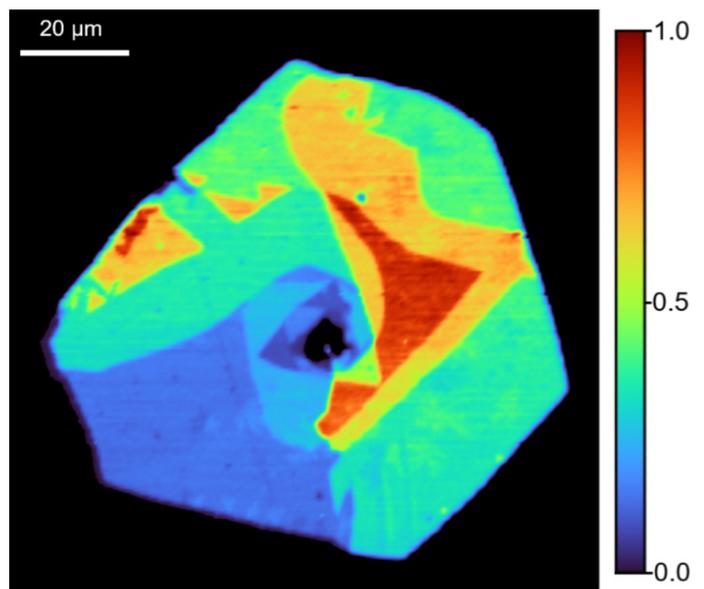
FIGURE 4. Second harmonic generation imaging of a WS_2 flake grown using CVD. The SHG imaging reveals that the flake is a polycrystal with each crystalline domain having a different SHG response due to differing number of layers, stacking symmetries and growth orientations.

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