Strain-Induced Second Harmonic Generation Enhancement in Monolayer MoS_2 Flakes Edge

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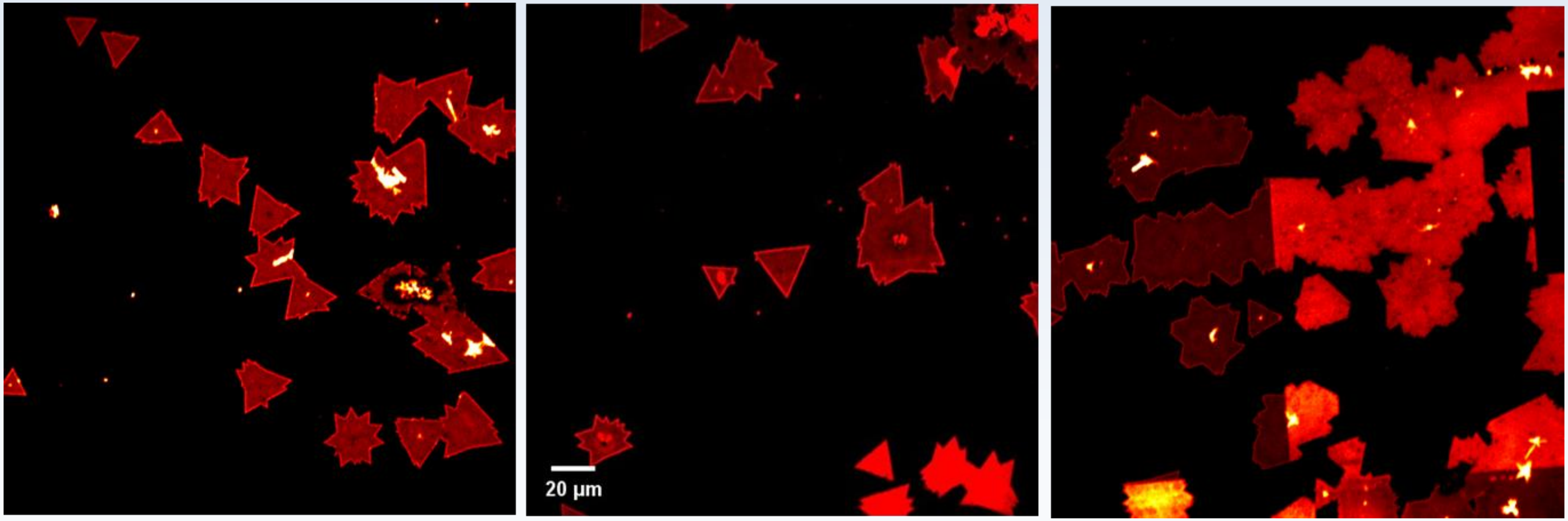
Motivation

Second harmonic generation (SHG) intensity mapping using laser scanning confocal microscope (LSCM)

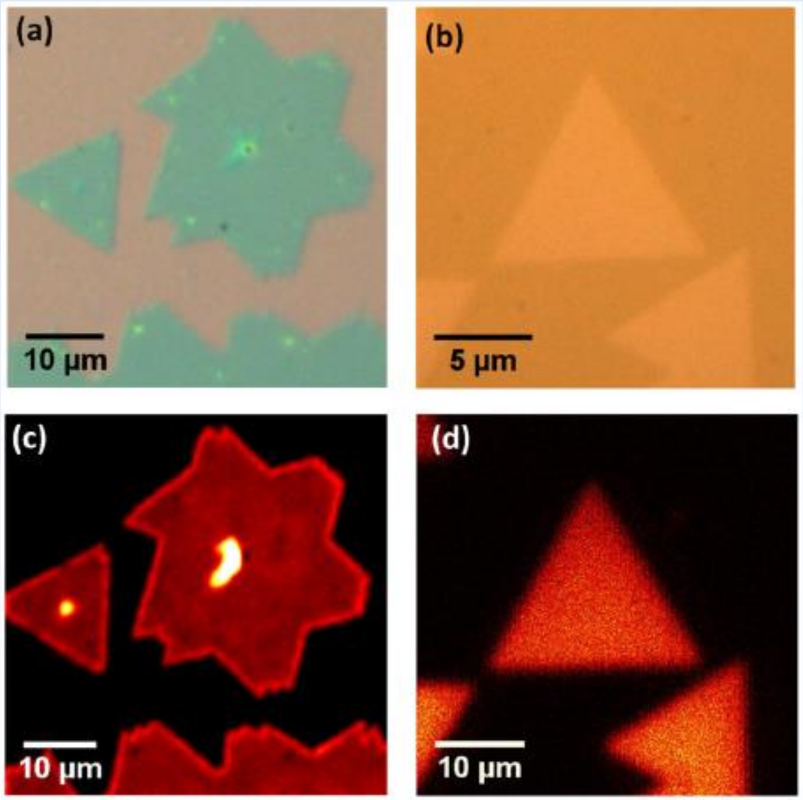


Sample: Monolayer MoS_2 grown on silicon substrate using CVD method

Abstract

- This edge enhanced SHG may be attributed to the intrinsic strain effect induced during the CVD growth condition, which modulates the nonlinear susceptibility.
- Micro-Raman spectroscopy analysis shows a blue shift in A_{1g} phonon mode at the flake edge, implies that the flake edge has a compressed and stiffened structure in an out-of-plane direction.
- The AFM analysis of the MoS_2 flake edge with enhanced SHG, indeed, detected a topographic height change as compared to the center region.

SHG Intensity Mapping

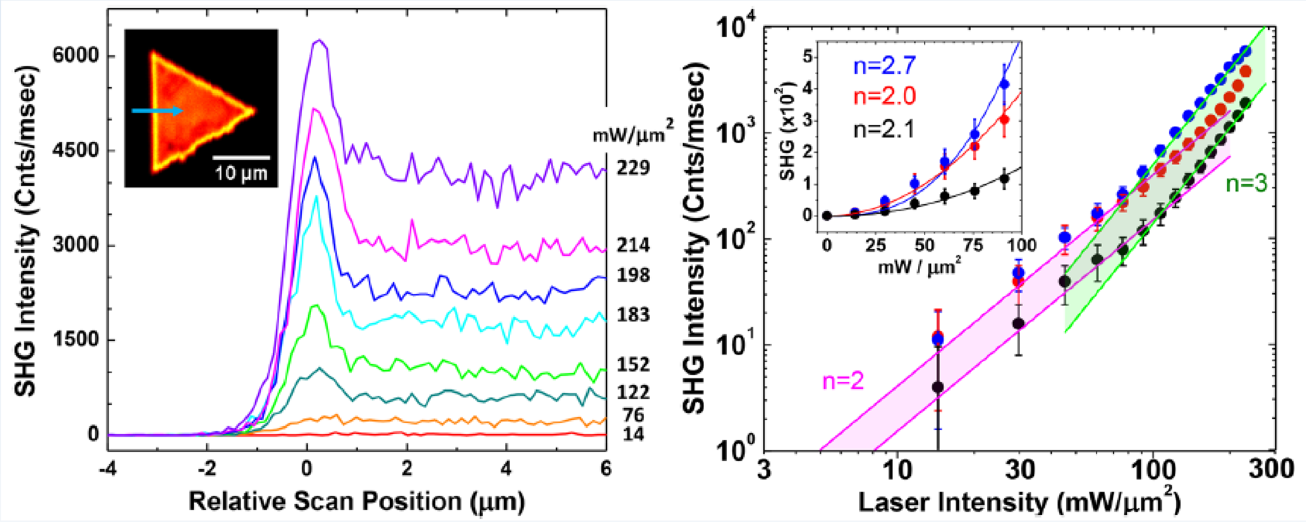


(a) $MoS_2@SiO_2$: Monolayer MoS_2 grown on a silicon substrate.
(b) $MoS_2@Al_2O_3$: Monolayer MoS_2 grown on a sapphire substrate.

Selected SHG intensity images of (c) $MoS_2@SiO_2$ and (d) $MoS_2@Al_2O_3$ acquired using LSCM and 1064 nm femtosecond laser excitation.

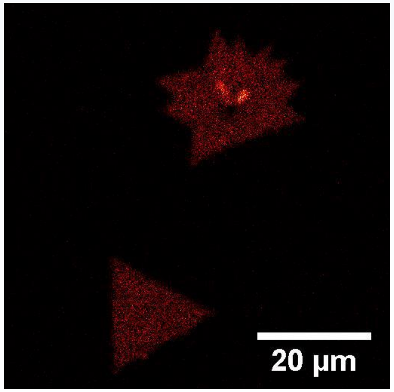
- MoS_2 @SiO_2 shows an enhanced SHG intensity near the flake edges. However, similar SHG edge enhancement is absent in MoS_2 @Al_2O_3.

Power Dependence



The power dependence of SHG intensity of MoS_2@SiO_2 center region, MoS_2@SiO_2 edge, and MoS_2@Al_2O_3. The inset figure shows the power dependence in low-intensity range and linear scale.

Red: MoS_2@SiO_2 flake center.
Blue: MoS_2@SiO_2 flake edge.
Black: MoS_2@Al_2O_3.



- The SHG contrast between the edge and the center became significant while increasing the laser intensity.
- A change in the gradient of the power dependence trend line can be observed as the laser intensity increases beyond 100 mW/μm². This may due to the laser-induced thermal strain on every regions of the sample.

Strain-Dependent SHG Intensity

- The strain dependent SHG intensity at the polarization direction ϕ is

$$I_{\square}^{(2)}(2\omega) \propto (A\cos(3\varphi - 3\delta) + B\cos(2\theta + \varphi - 3\delta))^2$$

with

$$A = (1-\nu)(p_1 + p_2)(\varepsilon_{xx} + \varepsilon_{yy}) + 2\chi_0^{(2)}$$

$$B = (1+\nu)(p_1 - p_2)(\varepsilon_{xx} - \varepsilon_{yy})$$

ν, Poisson's ratio

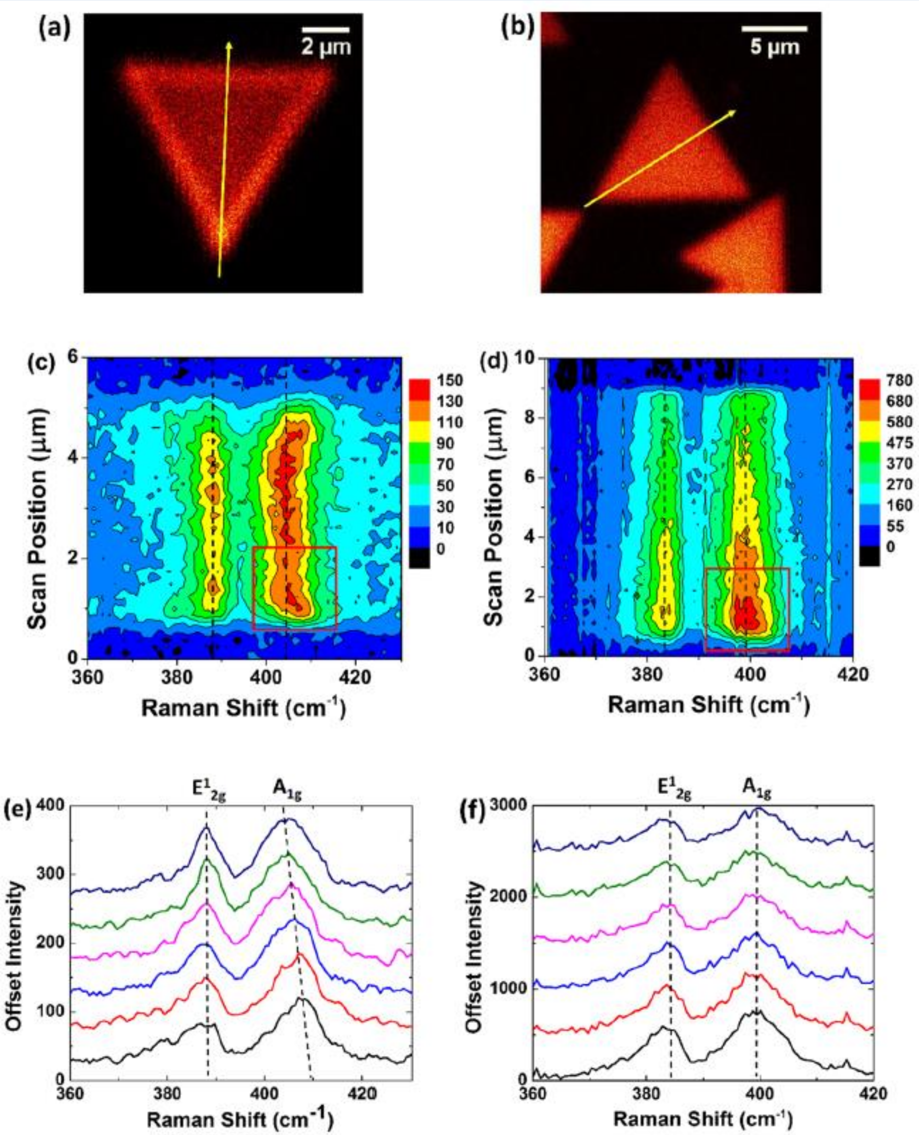
ε_{xx} & ε_{yy}, principal stresses

p_1 & p_2, free parameters in photoelastic tensor

$\chi_0^{(2)}$, nonlinear susceptibility

- The SHG variation with strain is related to the sum and difference of the photoelastic tensor elements p_1 and p_2.
- If the flake edge region is strained anisotropically, the asymmetry in principal strains will result in a nonzero term B, which further increases the SHG intensity.

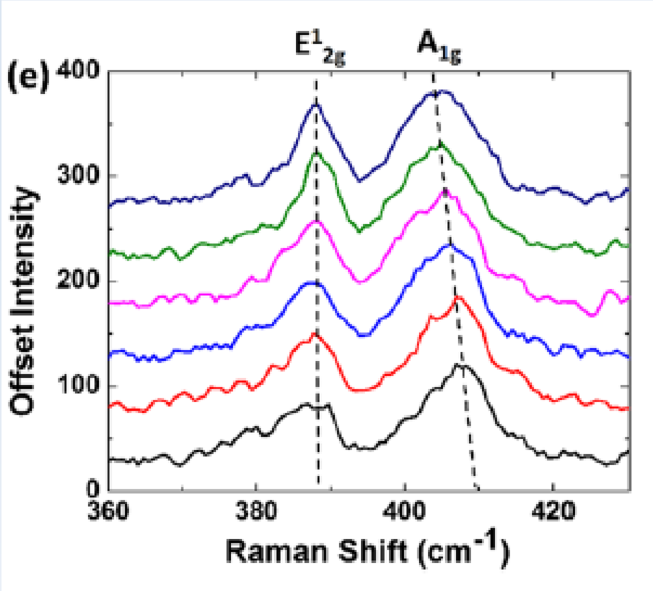
Micro-Raman Mapping

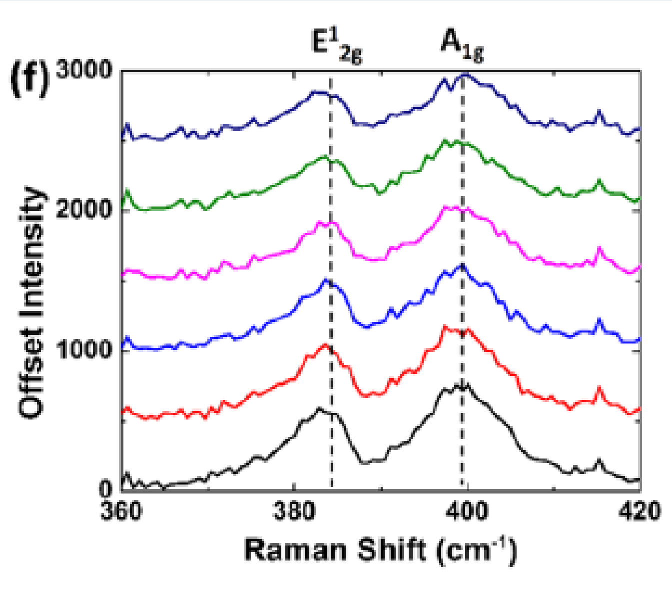


SHG intensity mapping of (a) MoS_2@SiO_2 and (b) MoS_2@Al_2O_3. The yellow arrows show the scanning direction of the line scan Raman spectra.

(c) The contour plot of the line scan Raman spectra in MoS_2@SiO_2. The scan position is following to the scanning direction in (a). (d) The contour plot of the line scans Raman spectra in MoS_2@Al_2O_3. The scan position is following to the scanning direction in (b).

(e) The Raman spectra at the edge regions [red box in (c)] of MoS_2@SiO_2. (f) The Raman spectra at the edge regions [red box in (d)] of MoS_2@Al_2O_3.

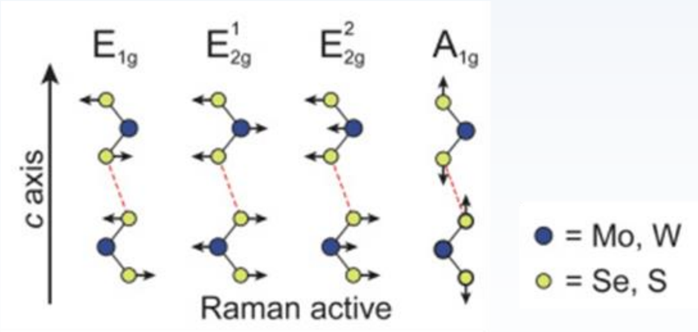




$MoS_2@SiO_2$ edge: Blue shift in out-of-plane A_{1g} phonon mode, from 404.9 to 407.7 cm^{-1} while the in plane E^1_{2g} phonon mode remains unchanged at 388 cm^{-1}.

$MoS_2@Al_2O_3$ edge: No shifting in both A_{1g} & E^1_{2g} phonon mode.

- The A_{1g} phonon mode at the flake center shows that it is in unstrained condition, indicates that the structure is compressed and stiffened in the out-of-plane direction at the flake edge.
- The SHG enhancement at the flake edge is related to this compressive strain. There is no SHG enhancement observed in sample $MoS_2@Al_2O_3$ because the strain is rather uniform throughout the sample.



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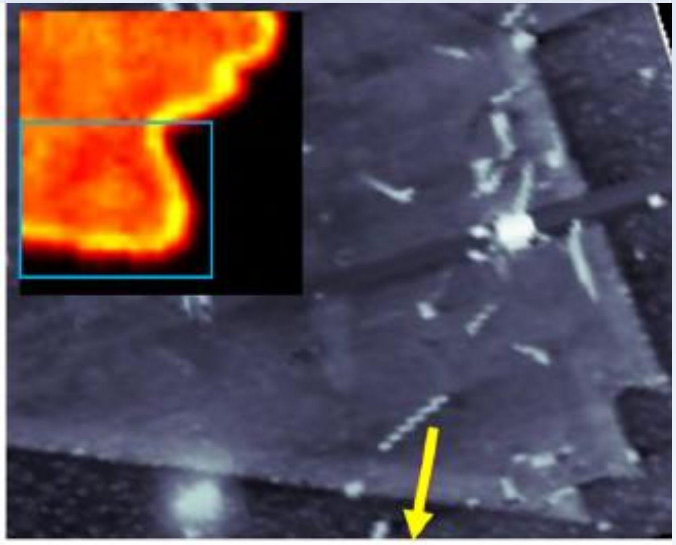
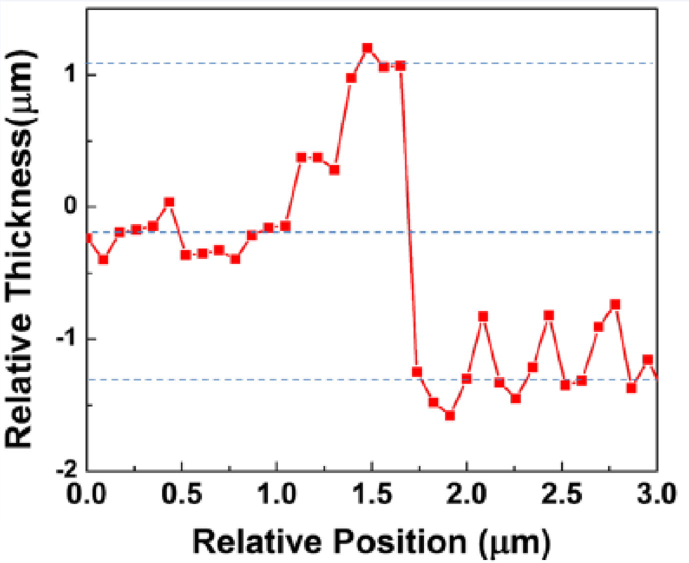
AFM Mapping



The AFM image of the $MoS_2@SiO_2$. The inset diagram shows the SHG image and the area of interest used in AFM analysis. The width of the white band top is ~0.34 μm.



The topography profile of the flake edge (yellow line). The flake edge has a thickness of ~1.25 nm larger than that in flake center region.

- The flake edge is probably a highly strained monolayer as compared to the flake center region.

Conclusion

- The AFM and Raman spectroscopy results show that the thickness and the phonon modes in the out-of-plane direction at the flake edge in those monolayer MoS_2 flakes are different from the flake center region.

- The intrinsic strain induced during the growth process could be the reason of the edge-enhanced SHG observed in MoS_2@SiO_2. The laser-induced thermal effect could further alter the power dependence behavior of the SHG signal from every sample.

- These results bring out the awareness of the residual strain effect on 2D TMD device performance and the adequate characterization of the strained structure by SHG intensity mapping.

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Thank you