The Origin of Strain-Enhanced Second Harmonic Generation in Monolayer MoS_2 Flakes

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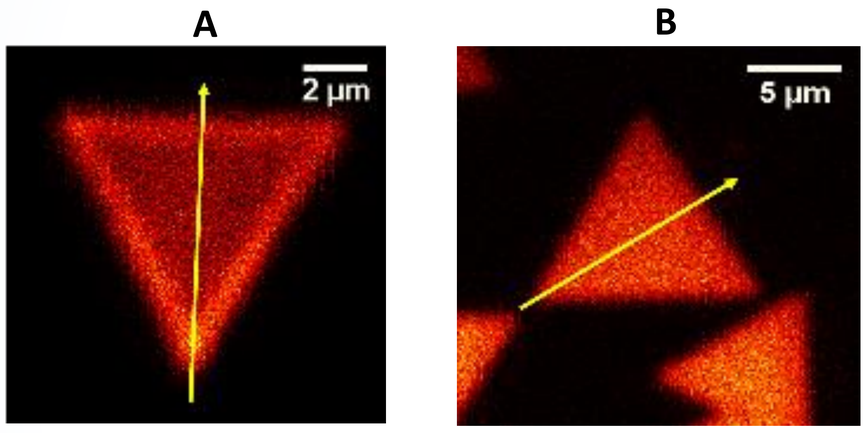
Abstract

In this work, we reported the observation of SHG enhancement at the edge of the monolayer MoS_2 flakes using a home-built laser scanning microscopy equipped with 1064 nm femtosecond laser light source and descanned SHG detection scheme. It is found that the SHG intensity at the edge region shows a cubic-power dependence, which may be attributed to the intrinsic strain effect induced in the edge region during the sample growth condition in CVD chamber. The AFM analysis of the MoS_2 flake with SHG enhancement phenomenon indeed detected a rather larger step height change at the edge as compared to the center region. Micro-Raman spectroscopy analysis using 532 nm laser shows a blue-shift in A_{1g} phonon mode at the flake edge, implies the flake edge has a compressed and stiffened structure in out-of-plane direction of the monolayer MoS_2 flake. We speculate that the aforementioned compressive strain at the flake edges could be the origin of the edge-enhanced SHG in certain MoS_2 flakes, where the local strain modulates the nonlinear susceptibility and leads the observation of edge-SHG enhancement in the edge of MoS_2 flake.

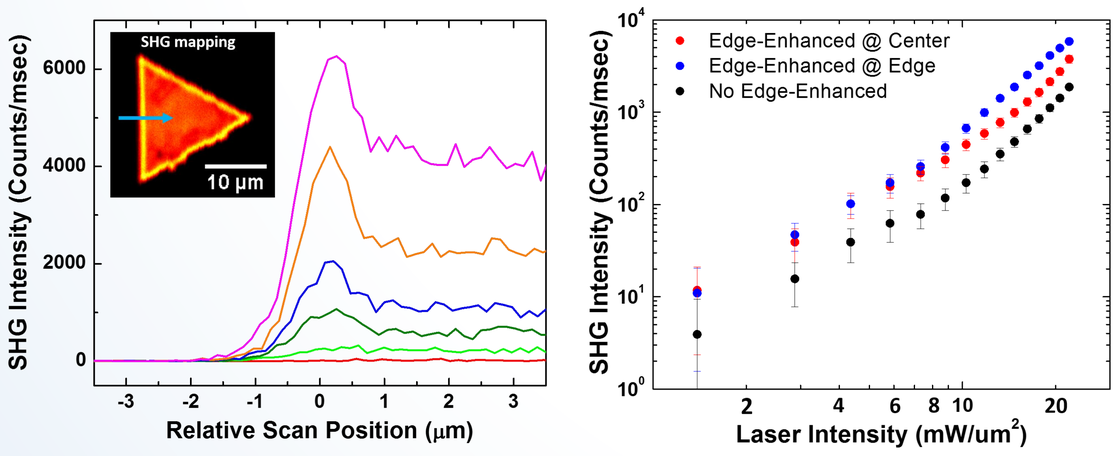
Sample Description

Sample	A	B
Fabrication	CVD-Grown	CVD-Grown
Quality	Low	High
Substrate	Silicon	Sapphire

SHG Intensity Imaging

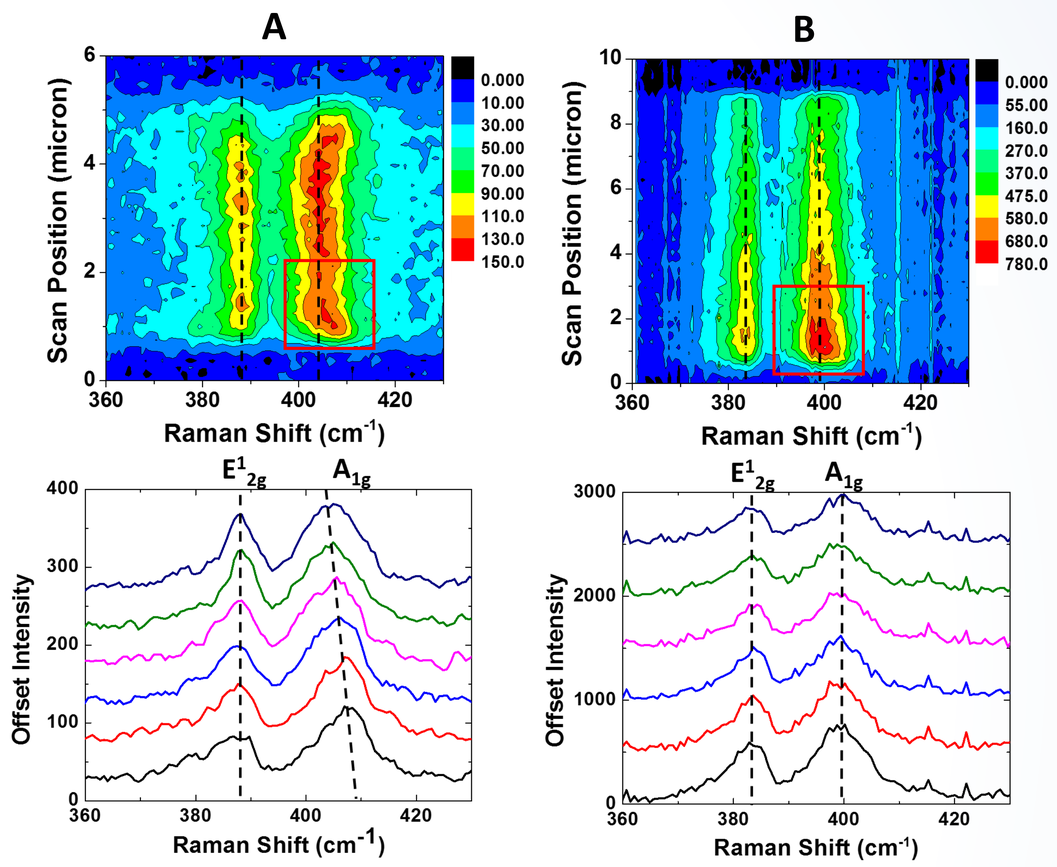


- SHG intensity imaging acquired using laser scanning microscopy and 1064 nm femtosecond laser excitation.
- The image on the left shows an edge-enhanced SHG, while the image on the right shows uniform SHG intensity.



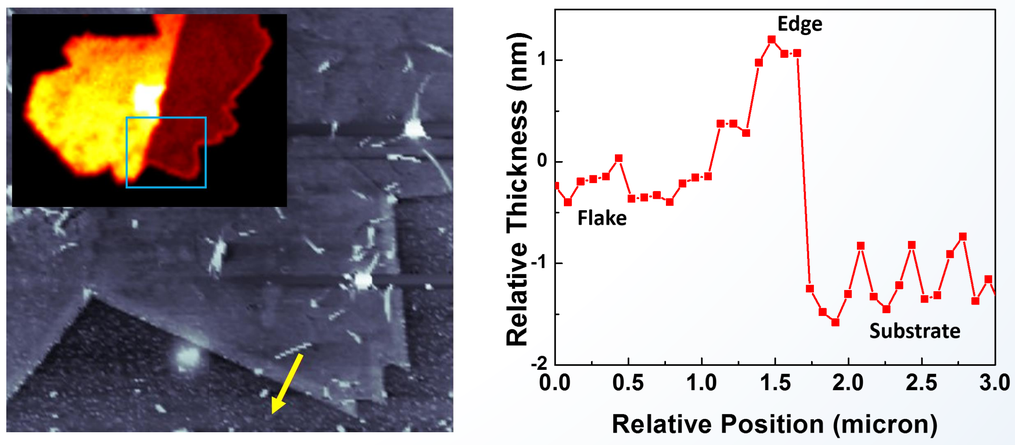
- The line scan SHG intensity across the edge of MoS_2 flake at various excitation laser intensity (increasing from bottom to top).
- The power dependence of SHG intensity at the edge and center region of both samples.
- A change in the gradient of the power dependence trend line can be observed as the laser intensity increased beyond 10 mW/μm2.
- The edge-enhanced SHG shows a cubic power dependence.

532 nm Raman Spectroscopy



- The contour plot of the line scan Raman spectra in both samples.
- The Raman spectra at the edge (red box) shows a blue-shifting for sample with edge-enhanced SHG.

AFM Results



- The AFM detected a rather larger step height change at the edge as compared to the center region (line profile along the yellow arrow).
- The flake edge has a thickness of approximately 1.25 nm compare to the flake center region.
- The width of the white band in the AFM image (left) is approximately 0.34 μm.

Conclusion

- The AFM and Raman spectroscopy results show that the thickness and the A_{1g} phonon mode at the flake edge in those monolayer MoS_2 flakes is different from the flake center region.
- This phenomenon is absent in the flake center region and high quality monolayer MoS_2 sample.
- The SHG intensity at the edge region shows a cubic-power dependence, which may be attributed to the intrinsic strain effect induced in the edge region during the sample growth condition in CVD chamber.
- We speculate that the intrinsic strain induced during fabrication process could be the reason of the edge-enhanced SHG.

Acknowledgement

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