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Effect of gold substrate on the excitonic properties of MoS_2: a final state sum frequency spectroscopy study

Tao Yang, Erick Pollmann, Stephan Sleziona, Marika Schleberger, R. Kramer Campen and Yujin Tong
Faculty of Physics, University of Duisburg-Essen, Lotharstr.1, 47057, Duisburg

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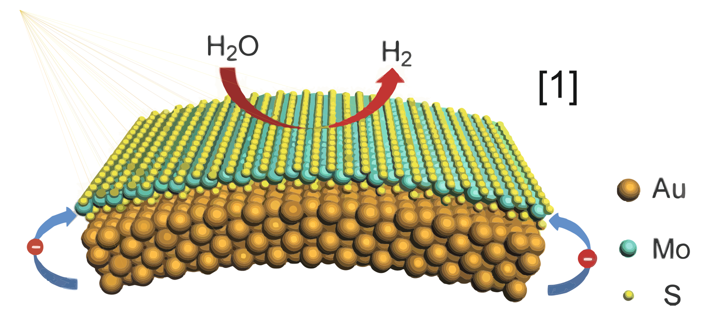
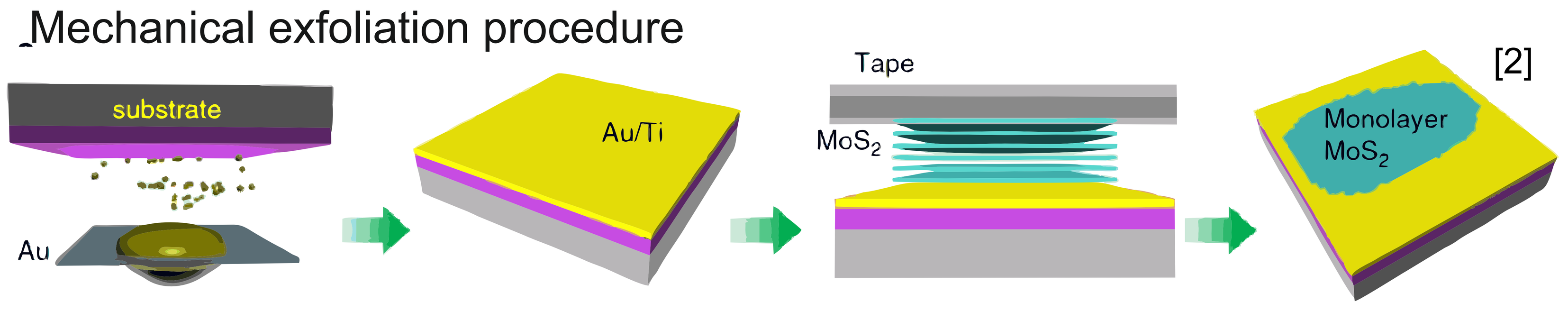
Introduction

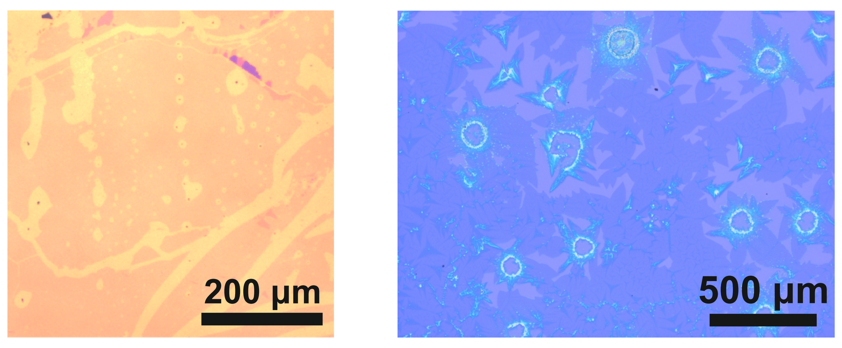
- Transition metal dichalcogenides (TMDCs) in both monolayer and few layer form are promising materials for application as optoelectronics, nonlinear optics and photocatalysts because of their direct bandgap in visible region of spectrum, large nonlinear optical response, pronounced activity for water splitting, and reasonable stability in ambient condition.
- Building devices with MoS_2 typically requires combining it with metals. Optimizing such devices thus requires understanding the metal-semicounductor interaction which determines the performance of these TMDCs devices. Gaining such understanding requires methods that are interface specific.
- **Sum frequency generation (SFG)** spectroscopy is a laser-based 2nd order nonlinear technique that can be applied to characterize the interfacial electronic structure and symmetry of optical response.



Sample preparation

Mechanical exfoliation procedure



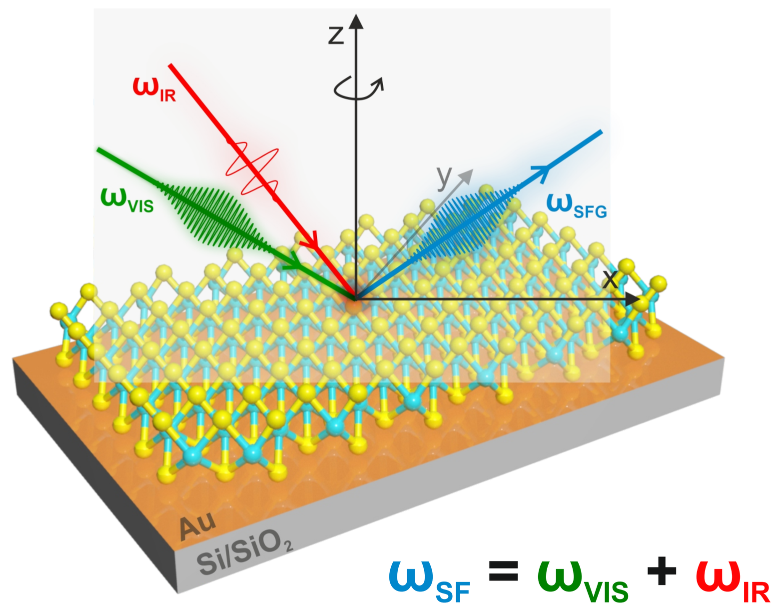


- Large flakes of monolayer MoS_2 can be obtained by both the mechanical exfoliation and chemical vapor deposition (CVD)!

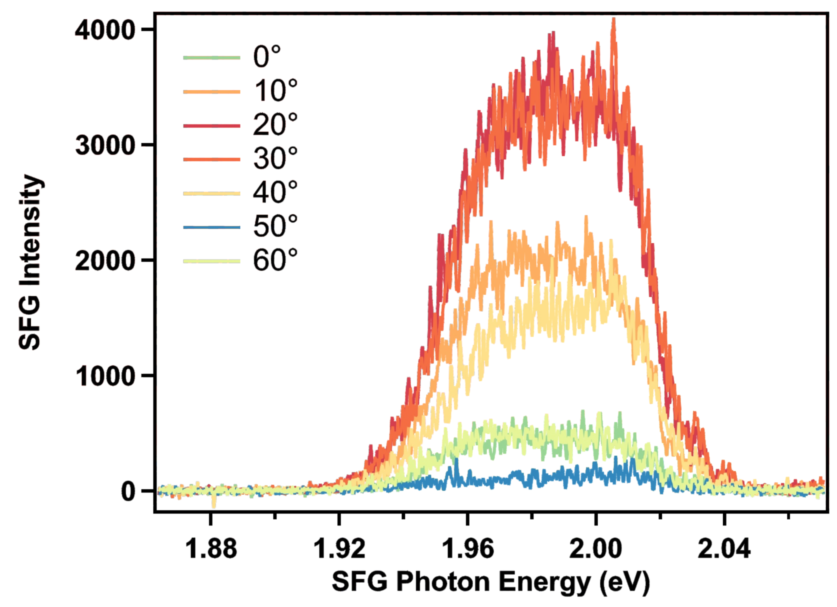
Optical images of exfoliated MoS_2 on gold and CVD MoS_2 on SiO_2

Method

Azimuthal dependent final state sum frequency spectroscopy



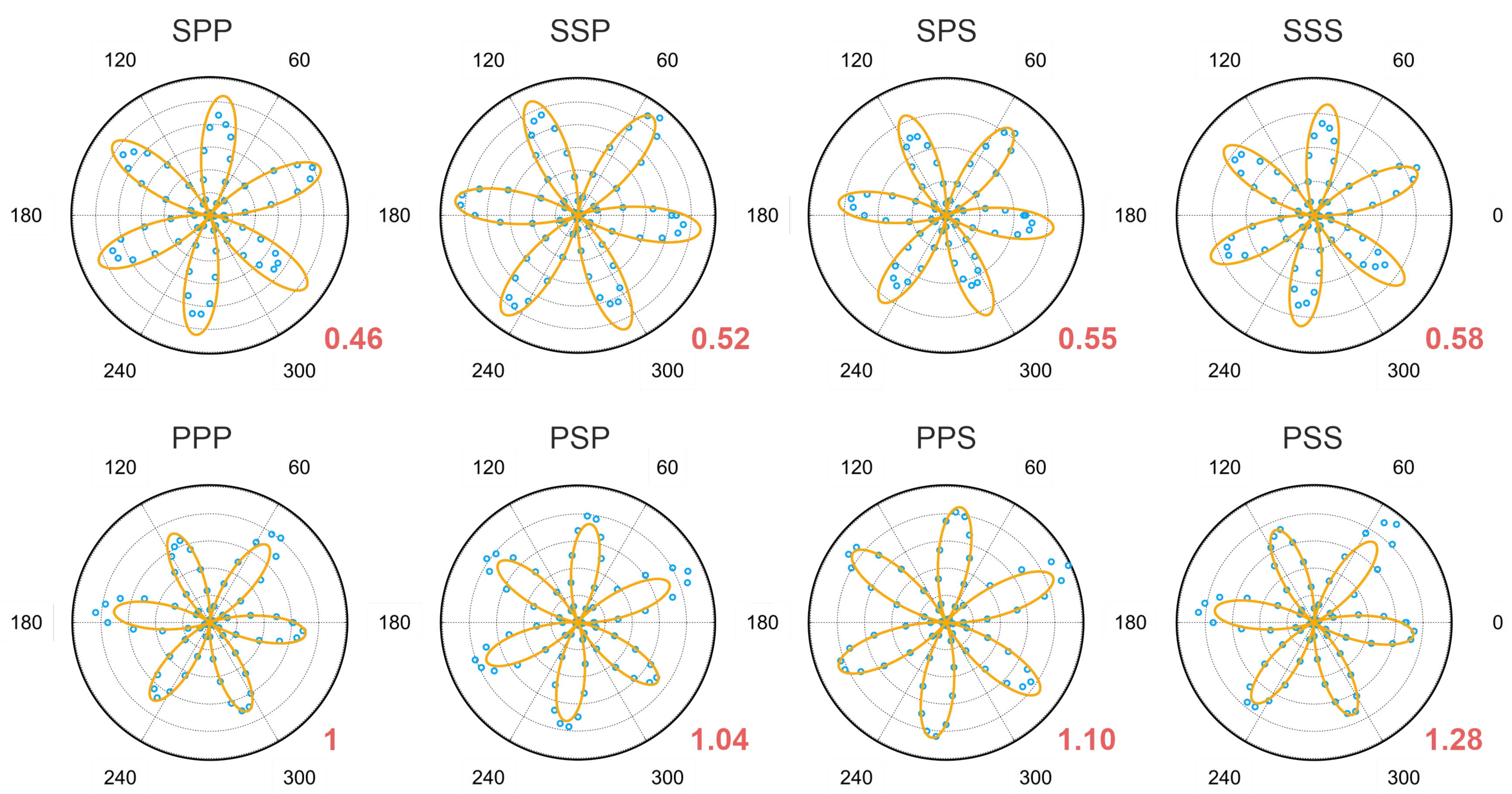
$\omega_{SF} = \omega_{VIS} + \omega_{IR}$



SFG spectra at different rotation angles

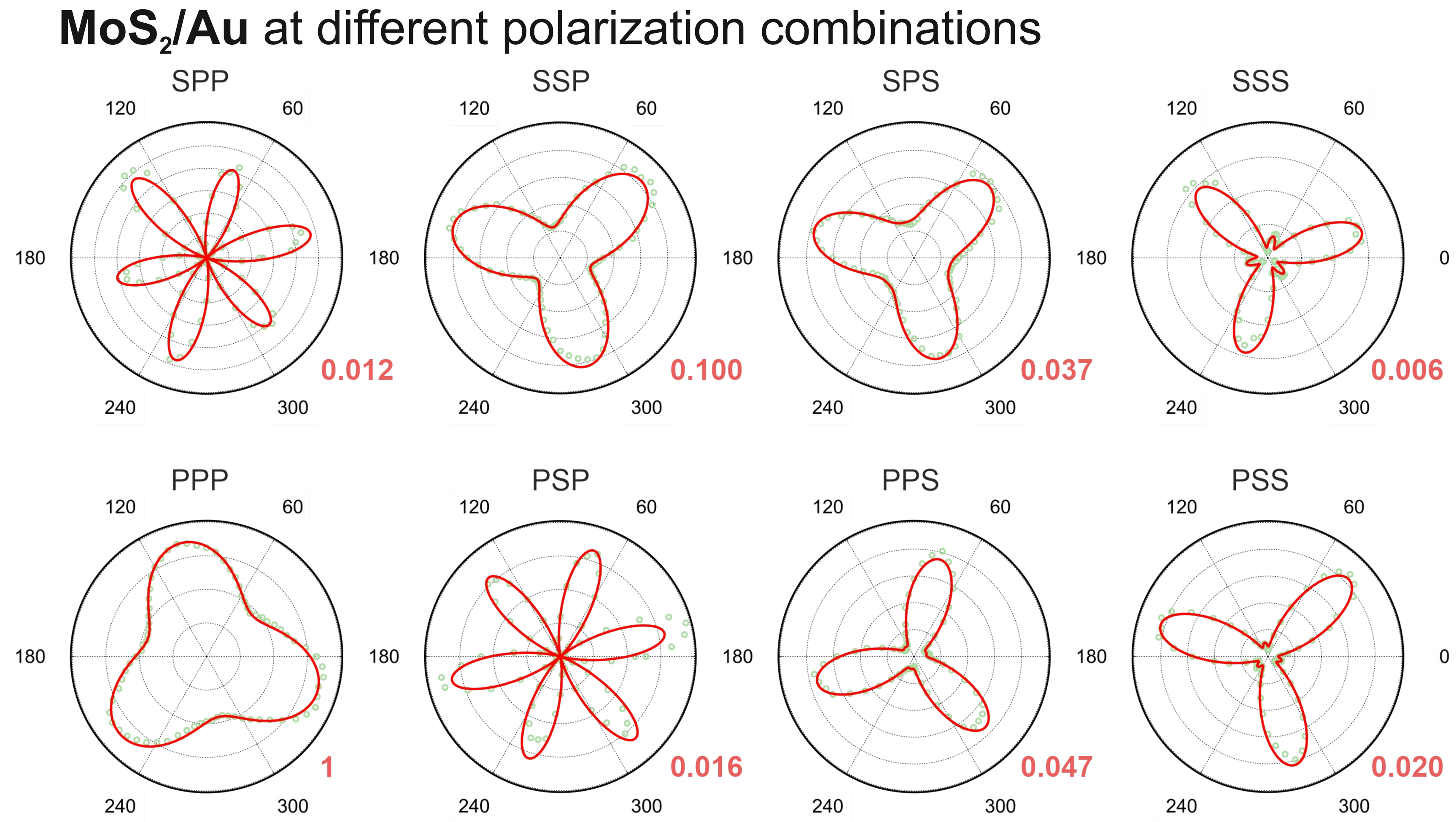
Azimuthal dependent results of MoS_2/SiO_2 and MoS_2/Au

MoS_2/SiO_2 at different polarization combinations

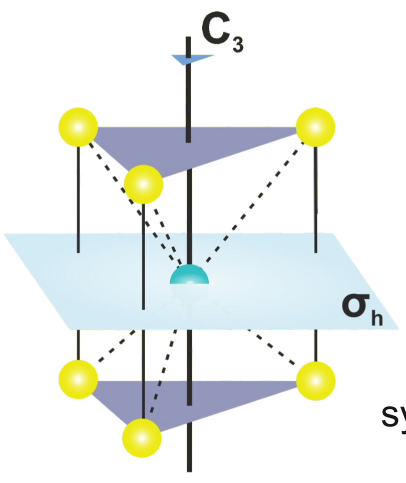


- All patterns show six-fold rotational symmetry
- The patterns can be divided into two groups based on the azimuth angles of the SFG maximum intensities
- The SFG intensities are polarization dependent

MoS_2/Au at different polarization combinations



- The patterns show dramatic difference compared to MoS_2 on SiO_2
- The three-fold symmetry patterns can be categorized into three types based on the shape and R (I_{max}/I_{min}) ratio
- The absolute maximum intensities vary significantly from each other



Description of SFG response

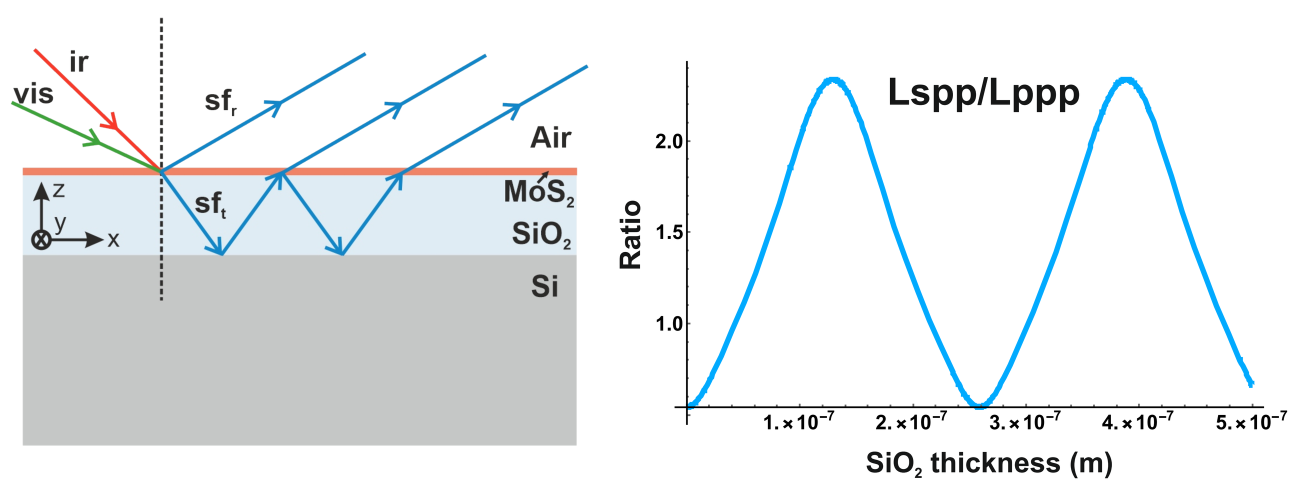
$$I_{sfg} \propto |\chi^{(2)}_{eff}|^2 I_{vis} I_{ir}$$

$$\chi^{(2)}_{eff} = [L_{sfg} \cdot \vec{e}_{sfg}] \cdot \chi^{(2)} : [L_{vis} \cdot \vec{e}_{vis}][L_{ir} \cdot \vec{e}_{ir}]$$

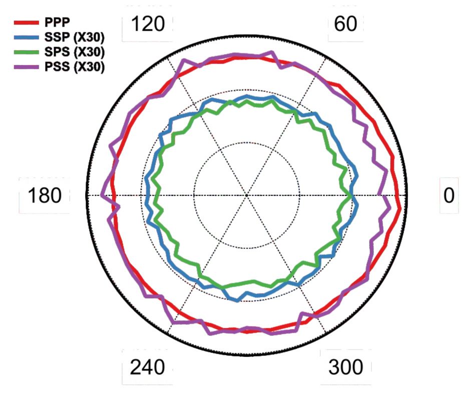
Point group of ML MoS_2 → D_{3h}

symmetry allowed component of the susceptibility tensor

$$\chi^{(2)}_{yyy} = -\chi^{(2)}_{yxx} = -\chi^{(2)}_{xxy} = -\chi^{(2)}_{xyx}$$

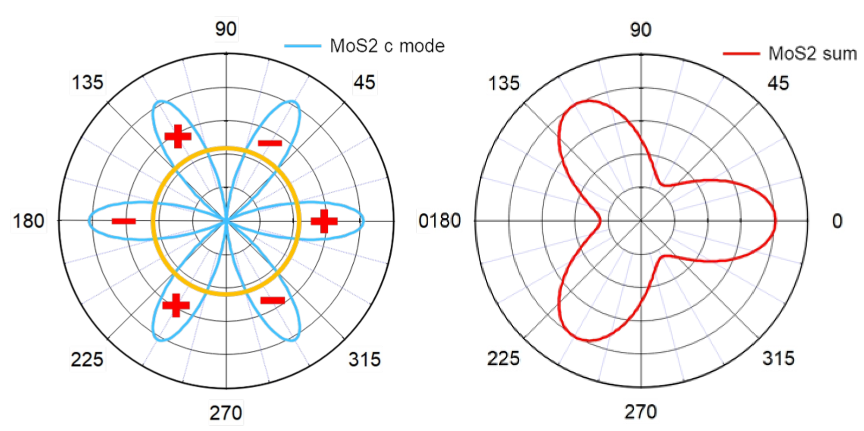


- The SFG intensity was manipulated by the thickness dependent interference
- The influence on the measured optical response from the dielectric substrate is negligible



- Three-fold symmetry is not the nature of PVD gold substrate

$$I_{SFG} \propto |\alpha + \beta \cos 3\theta|^2$$



Point group of ML MoS_2/Au → C_{3v}

$$\chi^{(2)}_{yyy} = -\chi^{(2)}_{yxx} = -\chi^{(2)}_{xxy} = -\chi^{(2)}_{xyx}$$

$$\chi^{(2)}_{xzx} = \chi^{(2)}_{yzy}, \chi^{(2)}_{xxz} = \chi^{(2)}_{yyz}, \chi^{(2)}_{zxx} = \chi^{(2)}_{zyy}, \chi^{(2)}_{zzz}$$

Point group of PVD Au → $C_{\infty v}$

C_{3v} ←?→ D_{3h} + $C_{\infty v}$

Sample	Azimuthal dependent	Azimuthal independent			
	χ_{yyy}	χ_{xxz}(ssp)	χ_{xzx}(sps)	χ_{zxx}(pss)	χ_{zzz}(ppp)
Au ($c_{\infty v}$)	-	28.9	3.1	2.6	-22.5
$MoS_2/SiO_2(D_{3h})$	1.0	-	-	-	-
$MoS_2/Au(C_{3v})$	2.2	49.6	3.3	-1.0	-13.0

$\chi^{(2)}_{yyy}(MoS_2/SiO_2)$=8*10^{-20} m^2/V $\chi^{(2)}_{yyy}(MoS_2/Au)$= 2*10^{-19} m^2/V

Conclusions

- Different laser polarization dependent symmetries have been observed on MoS_2/Au in comparison with that on dielectric substrate. These novel features can be exploited for the fabrication of nonlinear optoelectronics.
- The contribution to the SFG signal from the TMDC monolayer and that from the substrate can be well separated via azimuthal symmetry.
- The substrate-TMDC interaction can lift the symmetry of the optical response.

References

[1] Y. Tan, et al. Adv. Mater., 2014, 26, 8023–8028.
[2] Huang, Y, et al. Nat Commun, 11, 2453 (2020)