Anomalous Stokes–anti-Stokes ratio in $MoTe_2$ atomic layers

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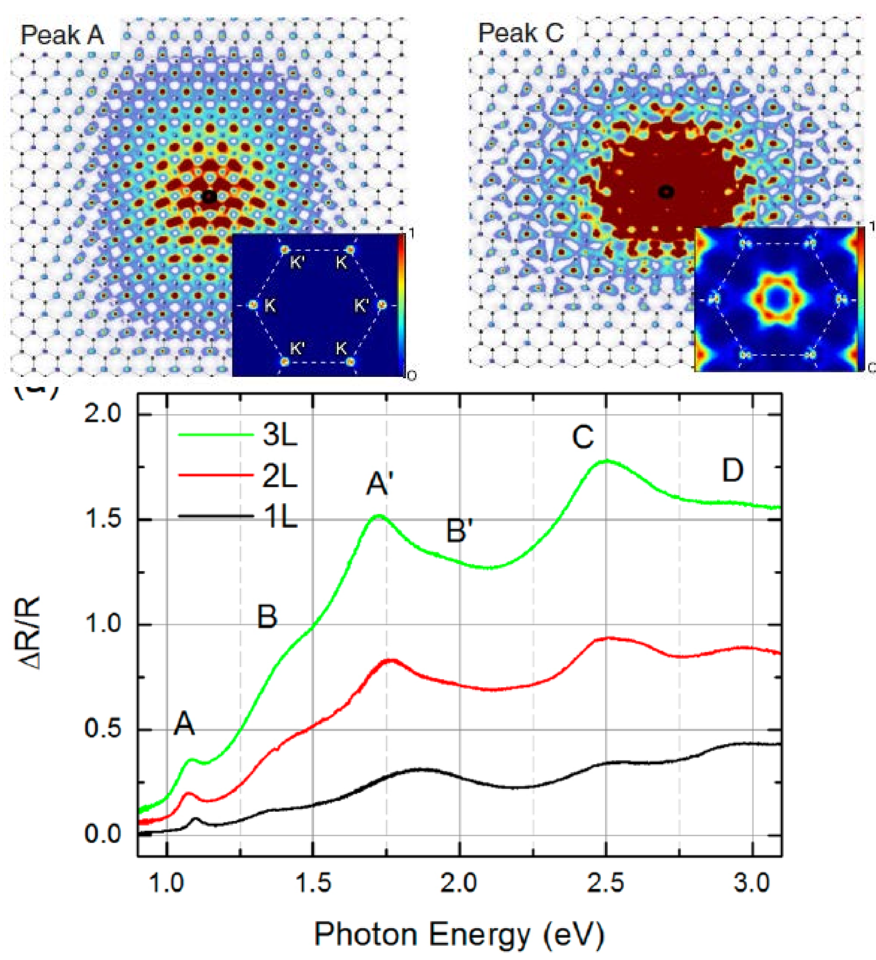
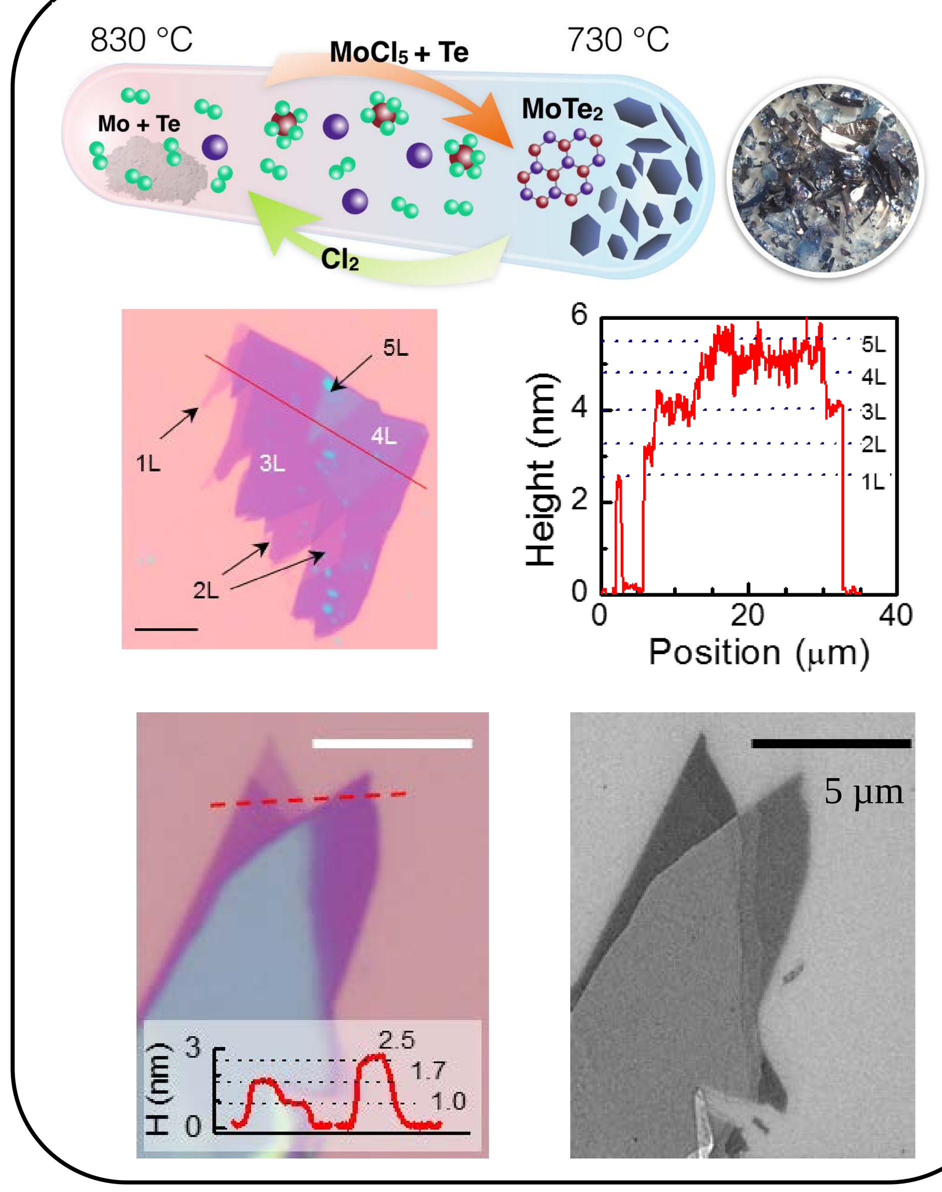
1. Motivation

Diverse excitons in TMDC



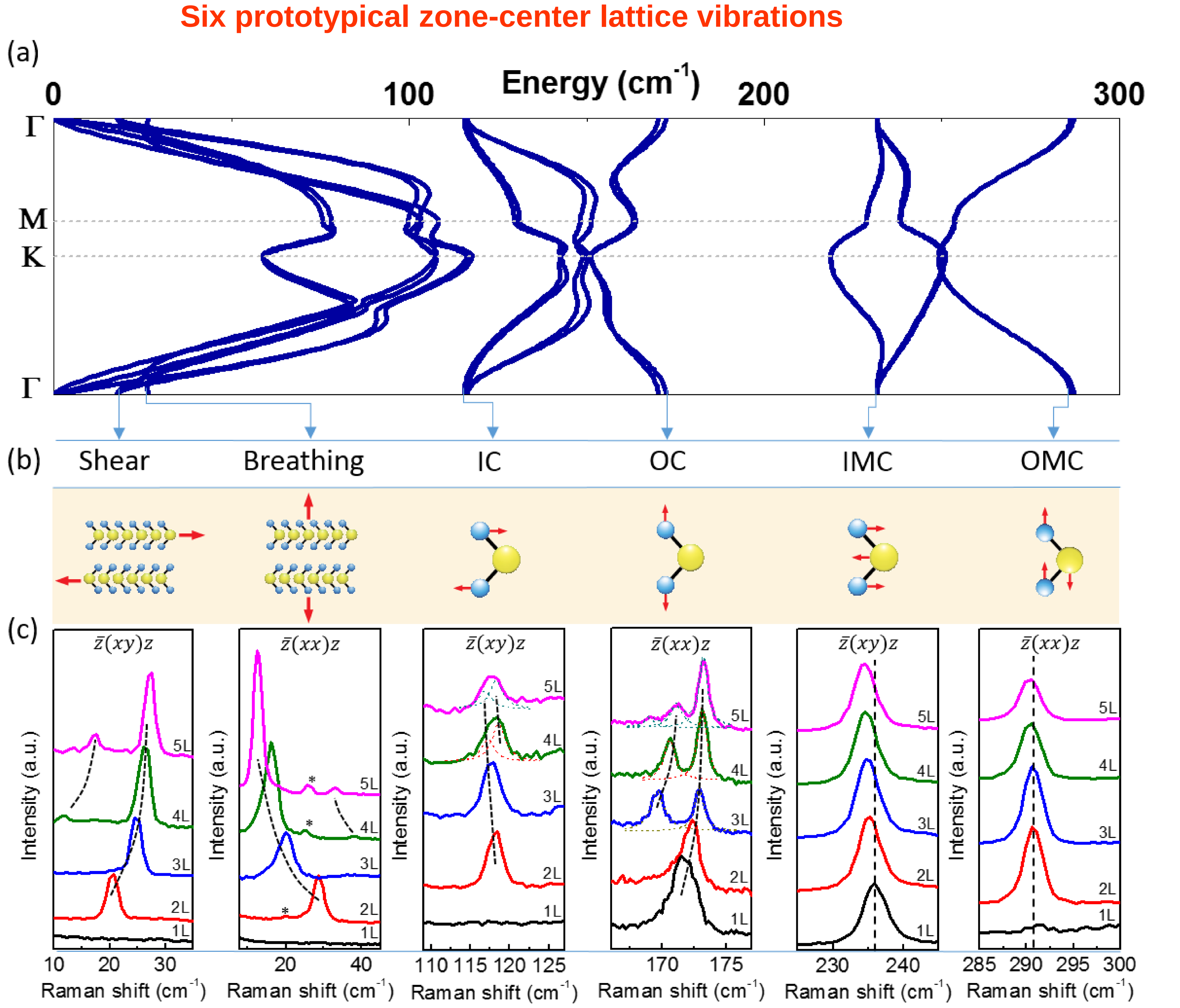
Qiu, Jornada and Louie Phys. Rev. Lett. 111, 216805 (2013).
Ruppert, Aslan and Heinz Nano Lett. 14, 6231 (2014).

2. Sample Preparation



3a. Raman Characterization

Six prototypical zone-center lattice vibrations



3b. Phonon symmetry

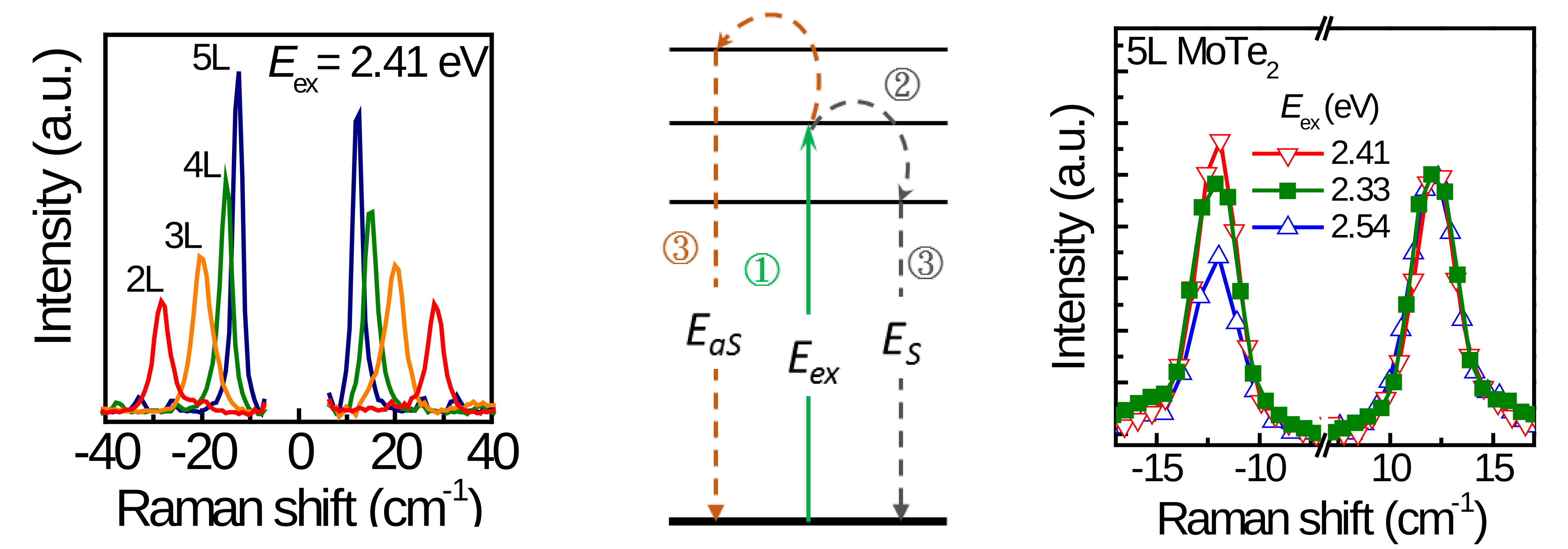
# of Layers	Sym. Grp.	σ_h / i Sym.	S	B	IC	OC	IMC	OMC
1	D_{3h}	+	-	-	-	1 A'_1	1 E'	-
		-	-	-	1 E''	-	-	1 A''_2
2	D_{3d}	+	1 E_g	1 A_{1g}	1 E_g	1 A_{1g}	1 E_g	1 A_{1g}
		-	-	-	1 E_u	1 A_{2u}	1 E_u	1 A_{2u}
3	D_{3h}	+	1 E'	1 A'_1	1 E'	2 A'_1	2 E'	1 A'_1
		-	1 E''	1 A''_2	2 E''	1 A''_2	1 E''	2 A''_2
4	D_{3d}	+	2 E_g	2 A_{1g}	2 E_g	2 A_{1g}	2 E_g	2 A_{1g}
		-	1 E_u	1 A_{2u}	2 E_u	2 A_{2u}	1 E_u	2 A_{2u}
5	D_{3h}	+	2 E'	2 A'_1	2 E'	3 A'_1	3 E'	2 A'_1
		-	2 E''	2 A''_2	3 E''	2 A''_2	2 E''	3 A''_2
bulk	D^4_{6h}	+	1 E_{2g}	1 B_{2g}	1 E_{1g}	1 A_{1g}	1 E_{2g}	1 B_{2g}
		-	-	-	1 E_{2u}	1 B_{1u}	1 E_{1u}	1 A_{2u}

Odd layers have mirror reflection symmetry σ_h.
Even layers and bulk have inversion symmetry i.

4a. Experimental Results I

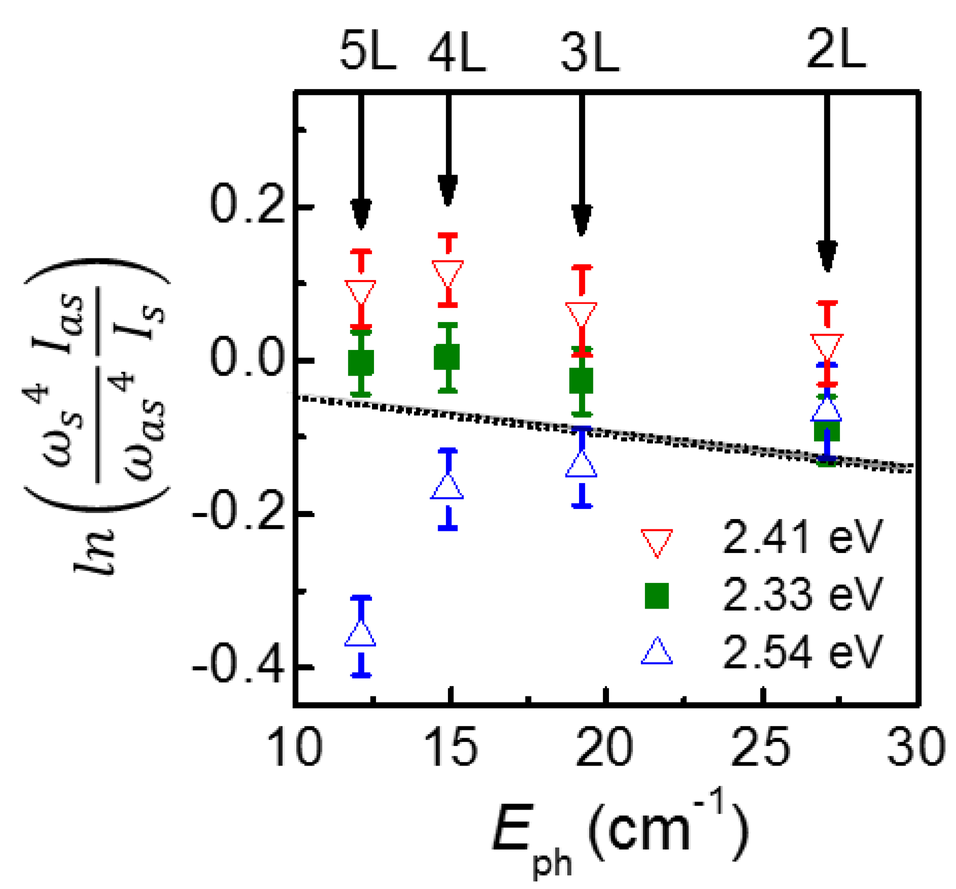
Anti-Stokes more intense than Stokes

Tunable Stokes–anti-Stokes ratio



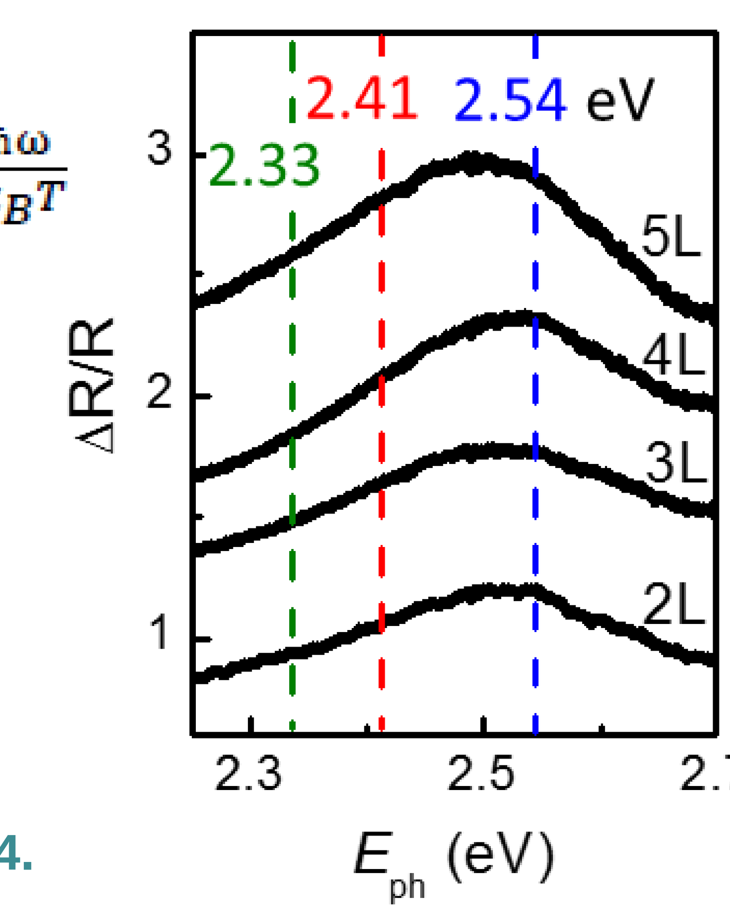
4b. Experimental Results II

C exciton resonance effect



$$\frac{I_{as}}{I_s} = R\frac{\omega_{as}{}^4}{\omega_s{}^4}\frac{n}{n+1} = R\frac{\omega_{as}{}^4}{\omega_s{}^4}e^{-\frac{\hbar\omega}{k_BT}}$$

$$\ln\left(\frac{\omega_s{}^4}{\omega_{as}{}^4}\frac{I_{as}}{I_s}\right) = \ln R - \frac{\hbar\omega}{k_BT}$$



arXiv:1603.03740
Scientific Reports (2016) 6: 28024.

5. Conclusions

- $MoTe_2$ atomic flakes are exfoliated from CVT grown crystals.
- Monolayers are readily distinguishable with OM, AFM and SEM.
- Raman scattering reveal all six types of zone center optical phonons.
- Intensity of the anti-Stokes breathing mode is tunable by laser exciton energy, and can be larger than the Stokes peak.
- The anomalous Stokes–anti-Stokes ratio can be explained by the resonance profile of the C exciton.
- The large anti-Stokes peak, more intense than the Stokes peak, creates a laser cooling channel in the transition metal dichalcogenide crystals.

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