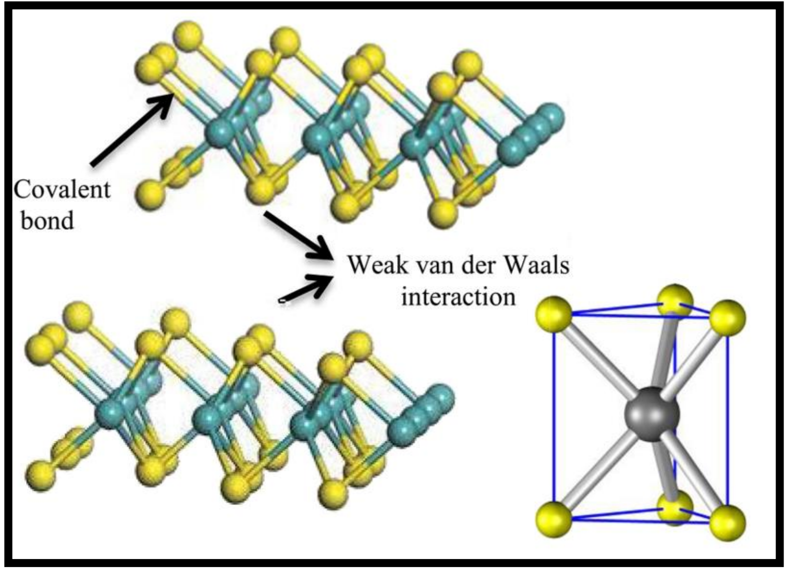
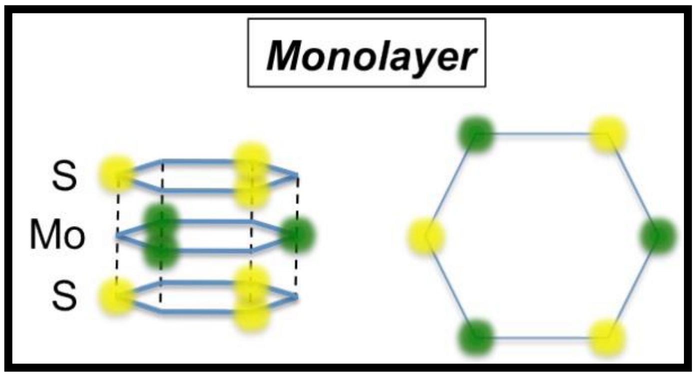
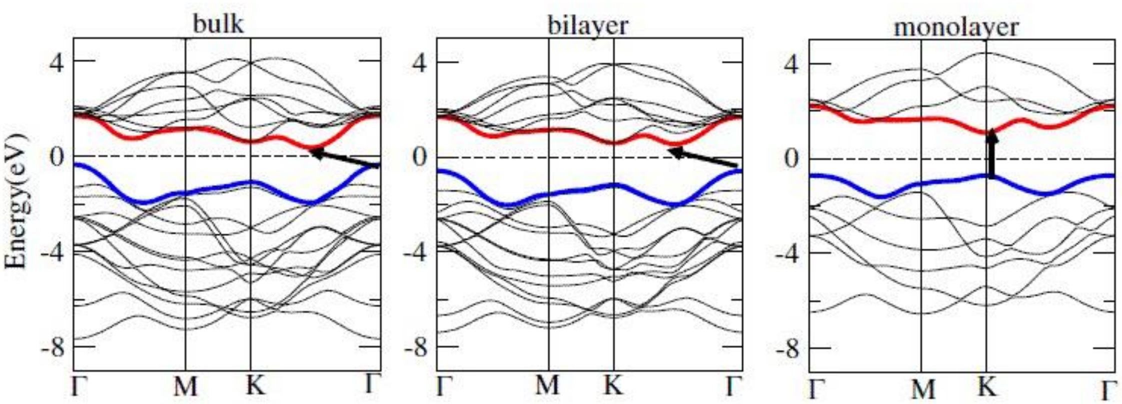
Intervalley dynamics in TMDs

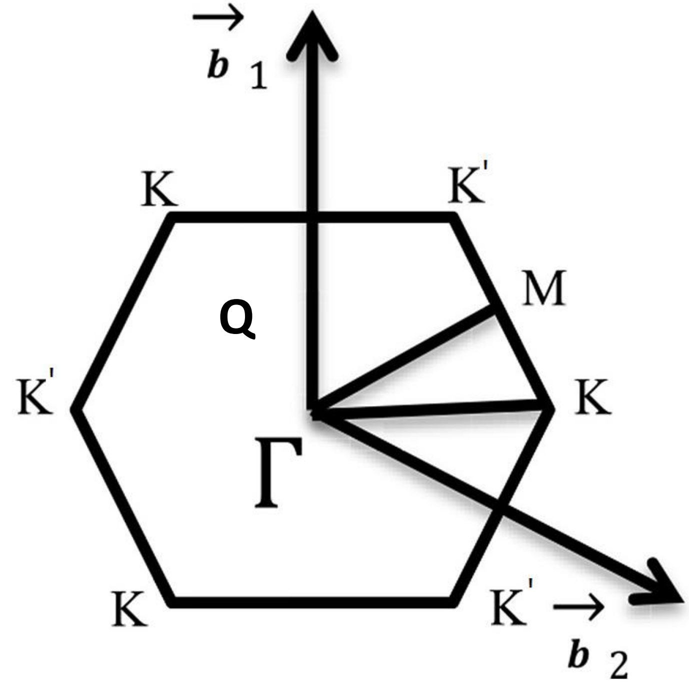
TMDs







Tawinan Cheiwchanchamnangij et al, Quasiparticle band structure calculation of monolayer, bilayer, and bulk MoS2

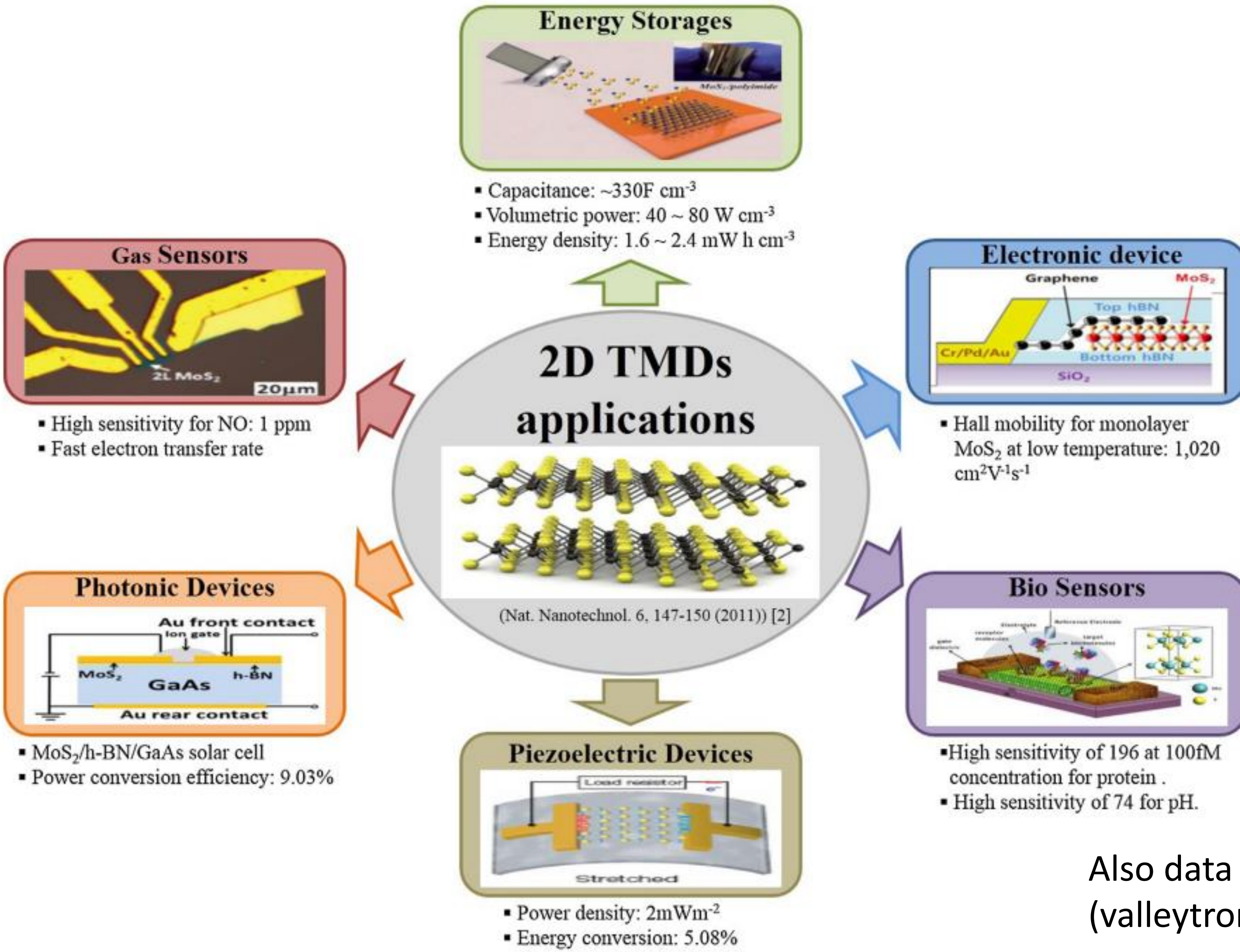


Generic TMD Brillouin Zone

Bilayer or thicker: indirect band gap between Γ and Q-point

Monolayer: girect band gap at the K (and K') point.

Technical applications



Also data storage (valleytronics)

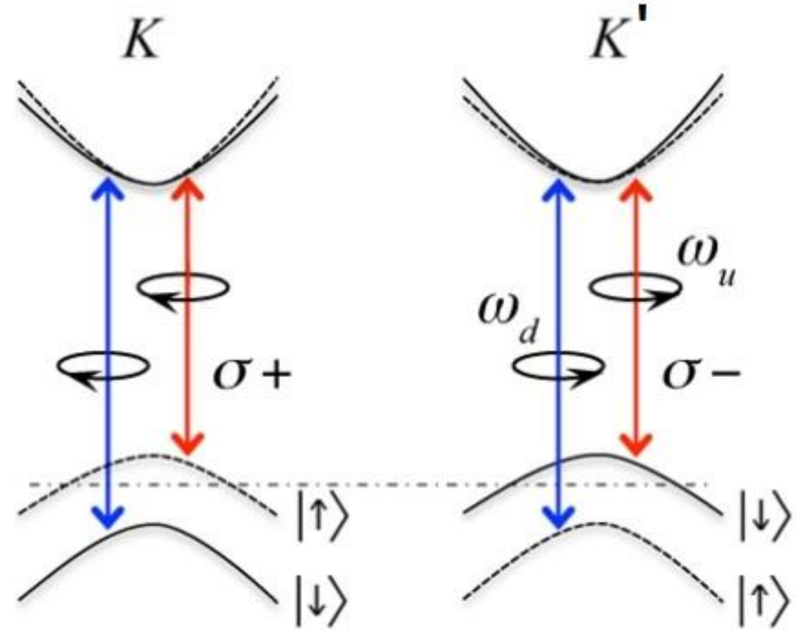
W. Choi et al, Recent development of two-dimensional transition metal dichalcogenides and their applications

Valley-pseudospin in 1L-TMDs

Monolayer: broken inversion symmetry

„Valley magnetic moment contribution from lattice structure leads to valley-dependent optical selection rules.“

Alexander V. Kolobov, Junji Tominaga
Two-Dimensional Transition-Metal dichalcogenides



D. Xiao et al, Coupled Spin and Valley Physics in Monolayers of MoS_2 and Other Group-VI Dichalcogenides

Short version: σ^{+} excites in the K-valley, σ^{-} excites in the K'-valley (linear excites in both)

ARTICLE

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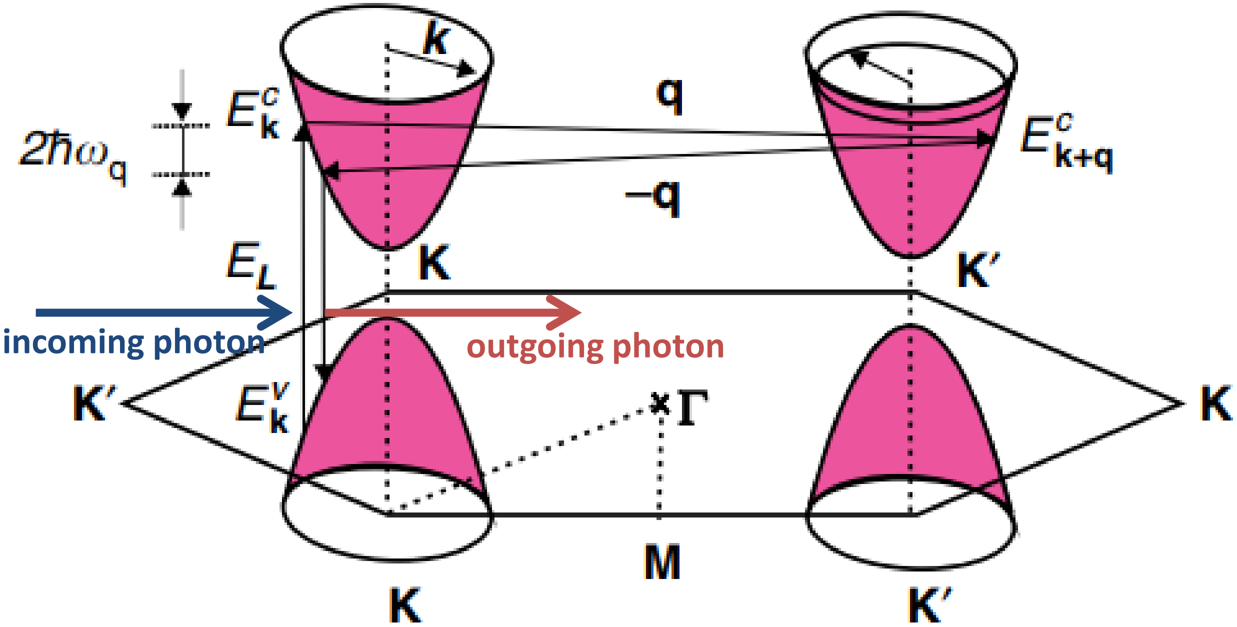
OPEN

Intervalley scattering by acoustic phonons in two-dimensional MoS_2 revealed by double-resonance Raman spectroscopy

Bruno R. Carvalho[1,*], Yuanxi Wang[2,*], Sandro Mignuzzi[3,4], Debdulal Roy[3,4], Mauricio Terrones[2,5,6], Cristiano Fantini[1], Vincent H. Crespi[2], Leandro M. Malard[1] & Marcos A. Pimenta[1]

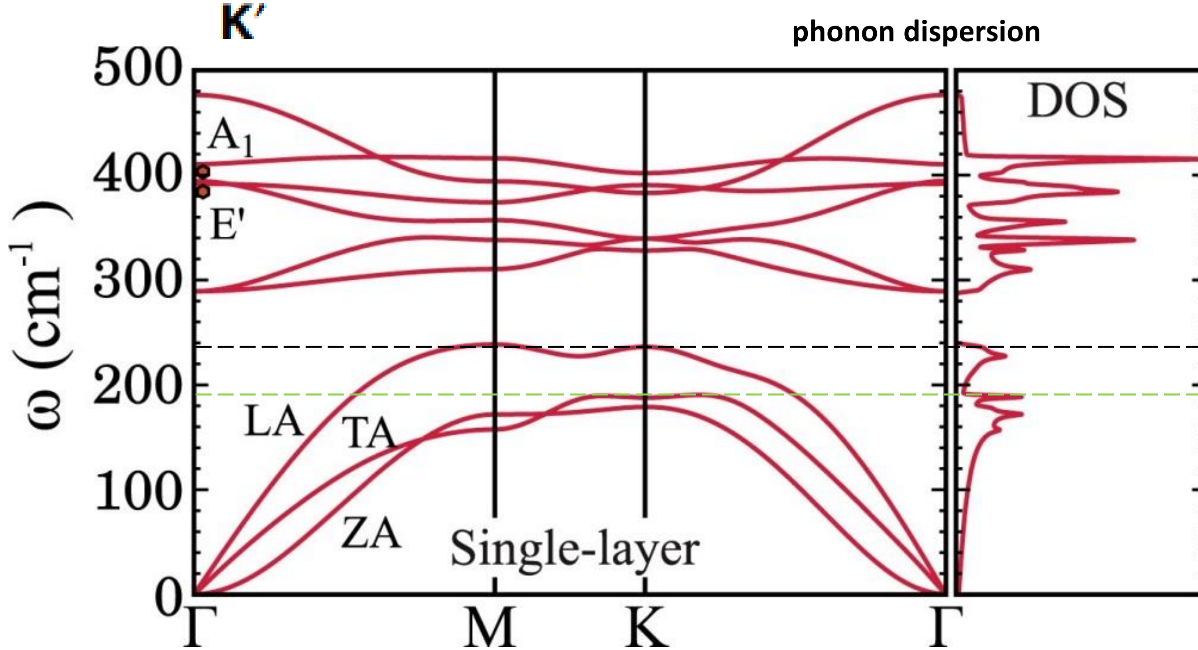
Double-resonance Raman scattering is a sensitive probe to study the electron-phonon scattering pathways in crystals. For semiconducting two-dimensional transition-metal dichalcogenides, the double-resonance Raman process involves different valleys and phonons in the Brillouin zone, and it has not yet been fully understood. Here we present a multiple energy excitation Raman study in conjunction with density functional theory calculations that unveil the double-resonance Raman scattering process in monolayer and bulk MoS_2. Results show that the frequency of some Raman features shifts when changing the excitation energy, and first-principle simulations confirm that such bands arise from distinct acoustic phonons, connecting different valley states. The double-resonance Raman process is affected by the indirect-to-direct bandgap transition, and a comparison of results in monolayer and bulk allows the assignment of each Raman feature near the **M** or **K** points of the Brillouin zone. Our work highlights the underlying physics of intervalley scattering of electrons by acoustic phonons, which is essential for valley depolarization in MoS_2.

Double resonant Raman scattering



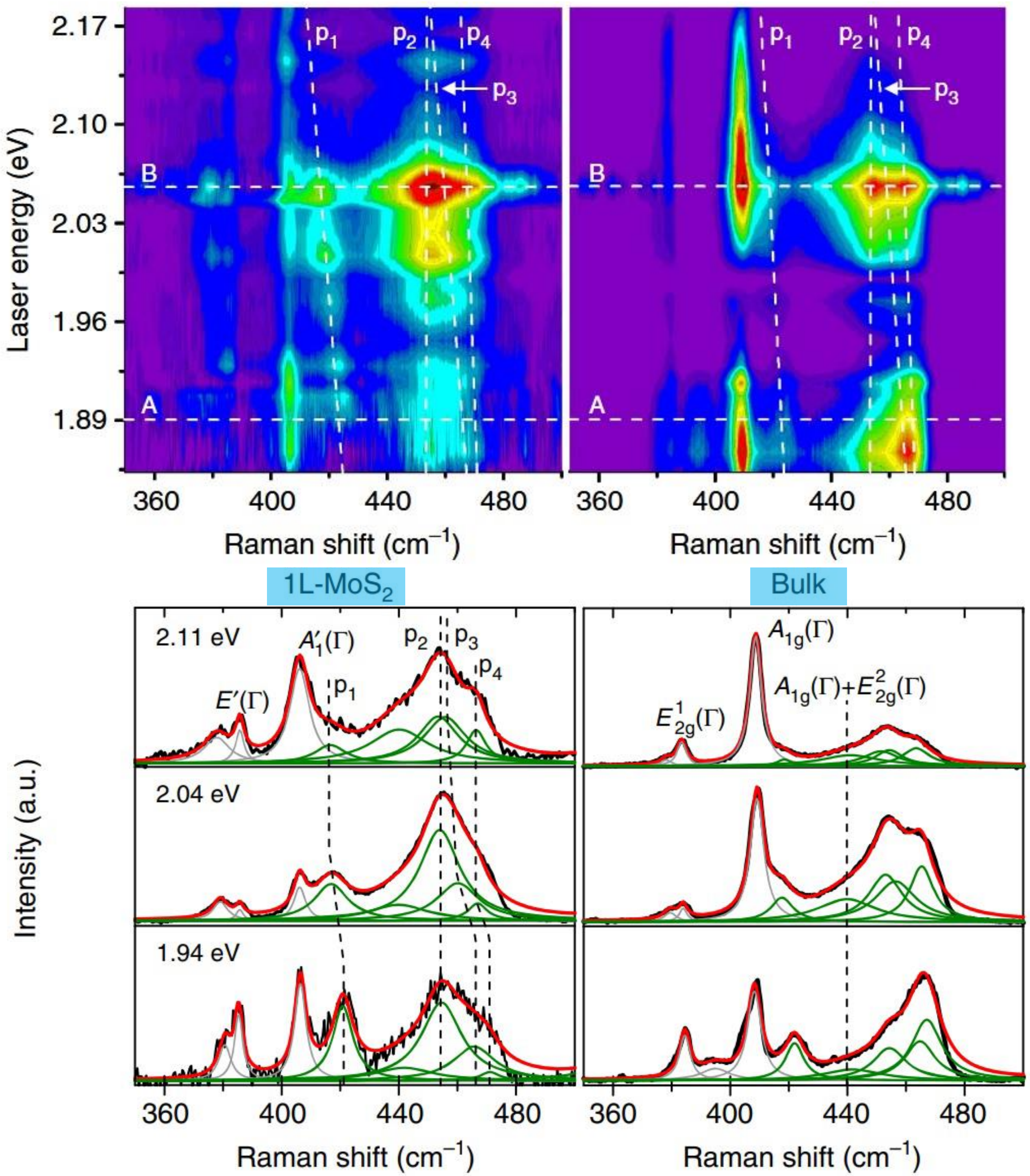
Photon excites electron to K-valley (1. resonance, band gap of ~1.6eV)

electron scatters to K'-valley via phonon (2. resonance, high DOS for phonons of required type) and back



A Molina-Sánchez et al, Phonons in single-layer and few-layer MoS_2 and WS_2

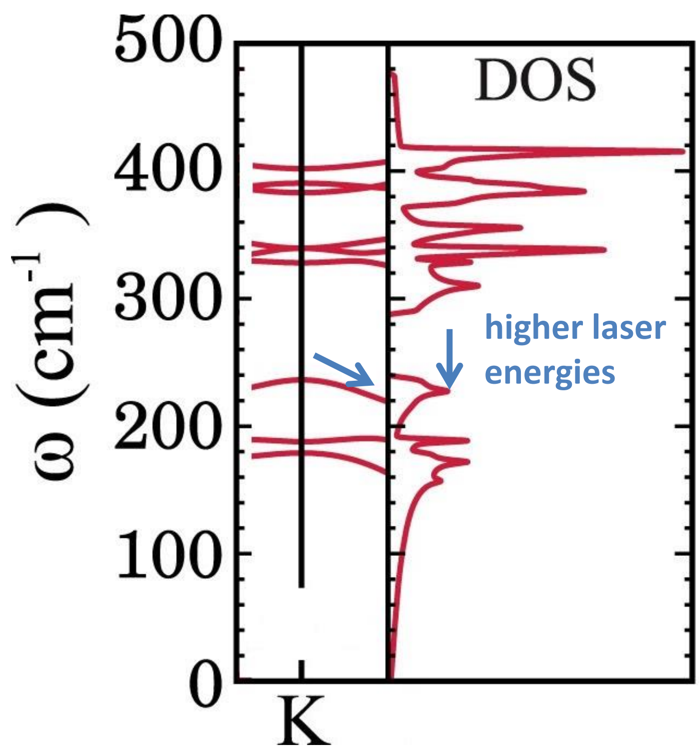
Frequency shift in two-phonon modes



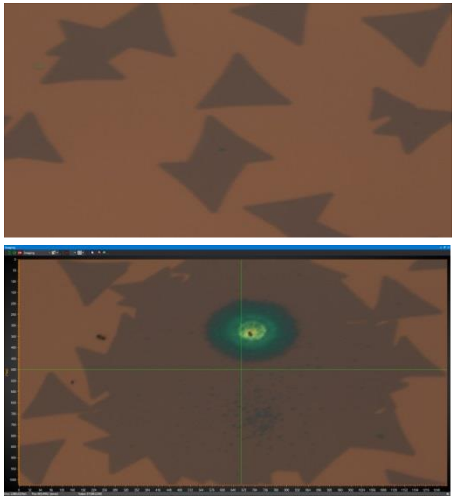
Peak at 460 cm-1 fitted by sum of Lorentzians

→ with higher excitation-energy, peaks shift to lower frequency

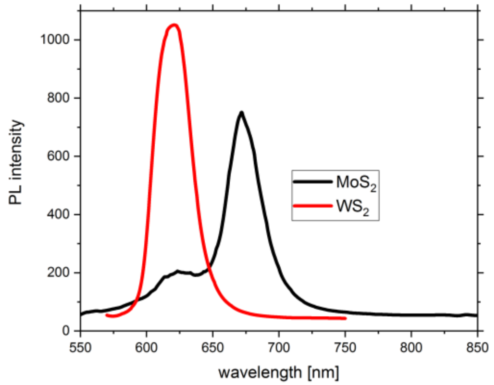
explained by intervalley scattering: Photons of higher energy couple to different electrons and phonons in 1.BZ



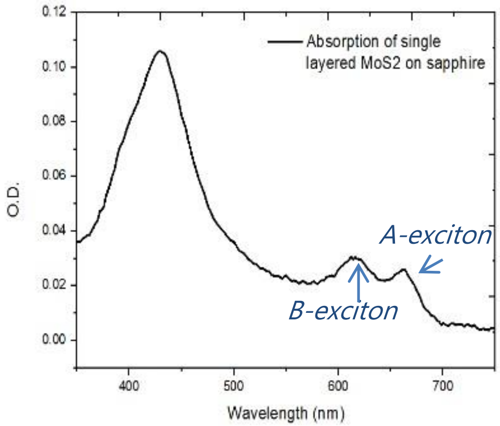
Sample characterisation



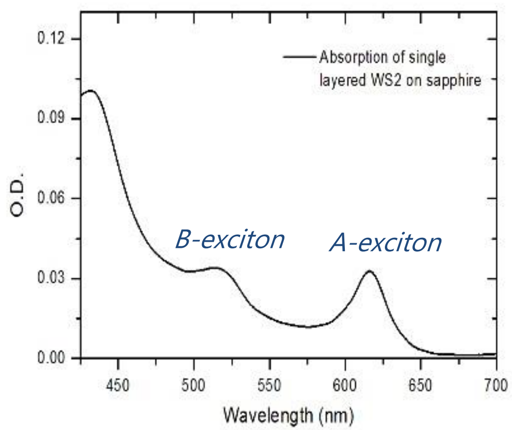
photoluminescence



absorption spectra

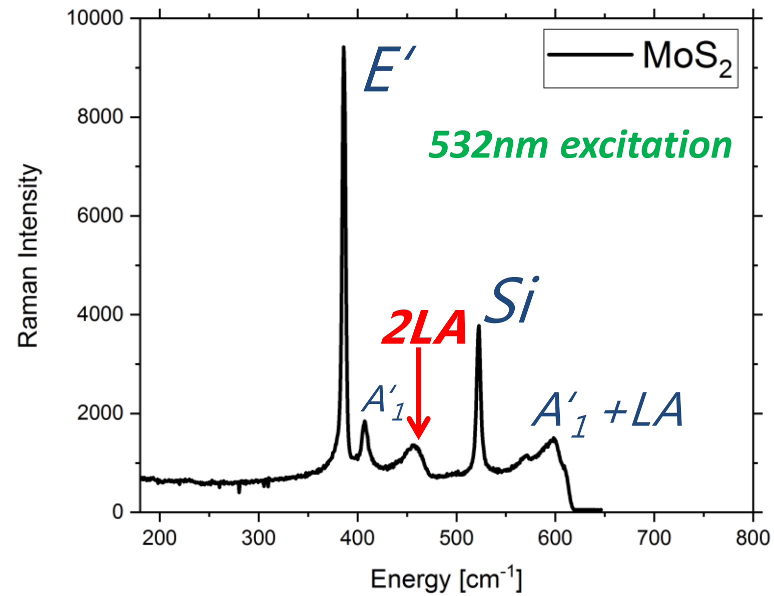


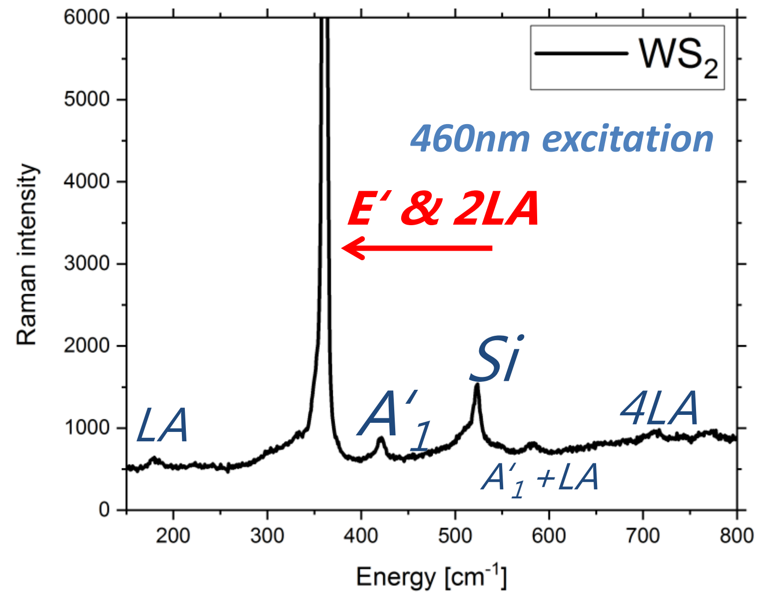
MoS_2



WS_2

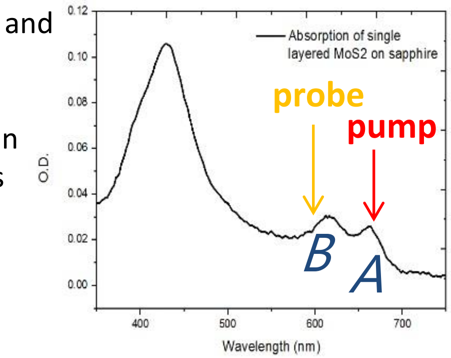
Raman spectra at T=300K, unpolarized

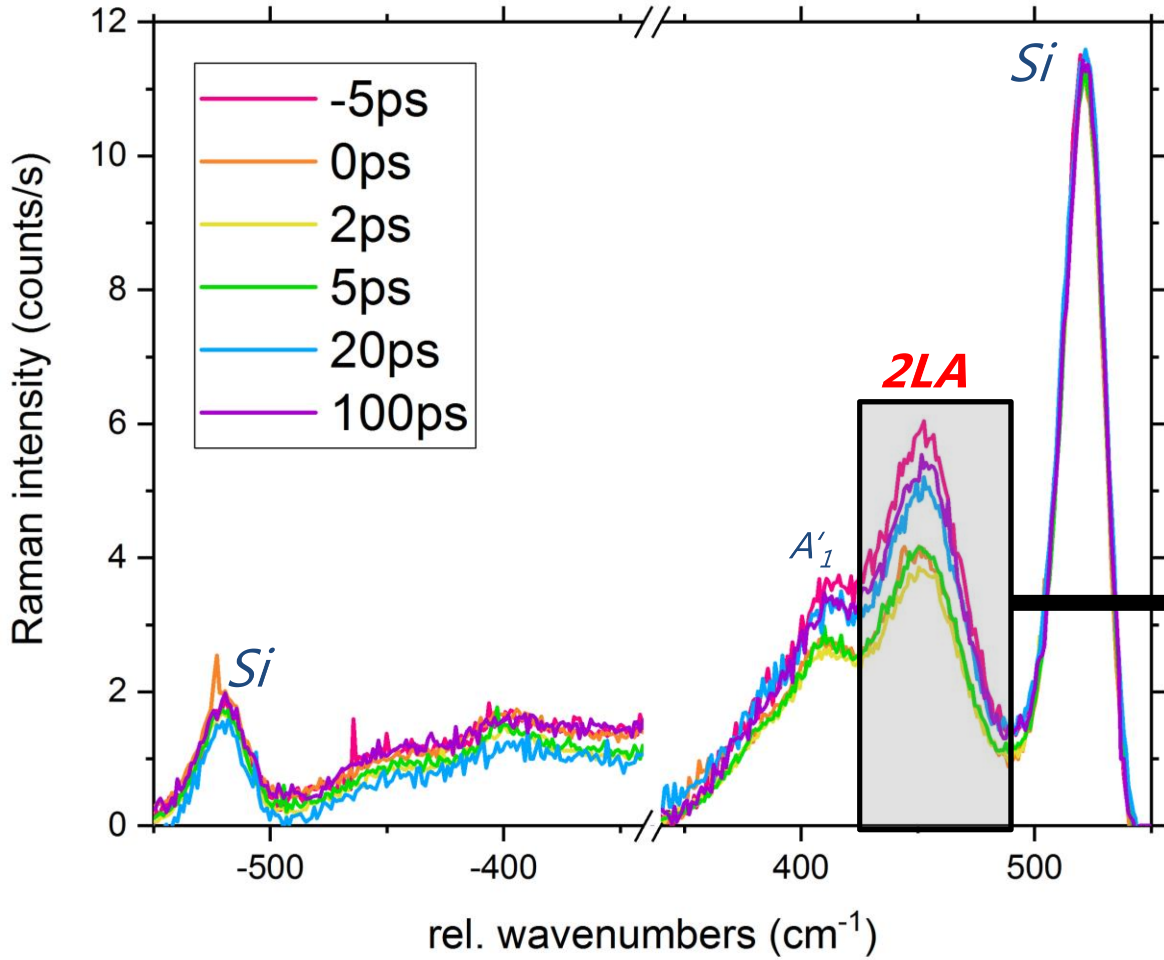


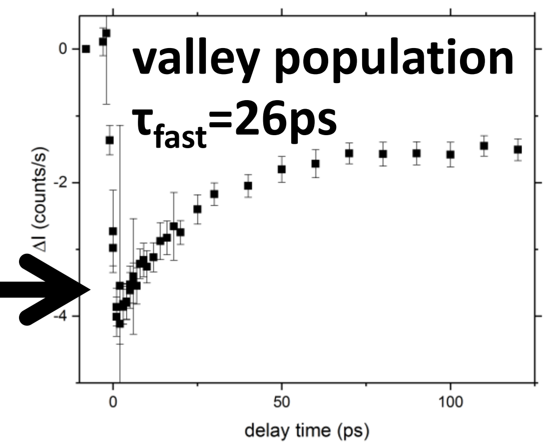


MoS_2 TiReRa

pump (650nm) and probe (605nm) with linear polarized light in excitonic bands





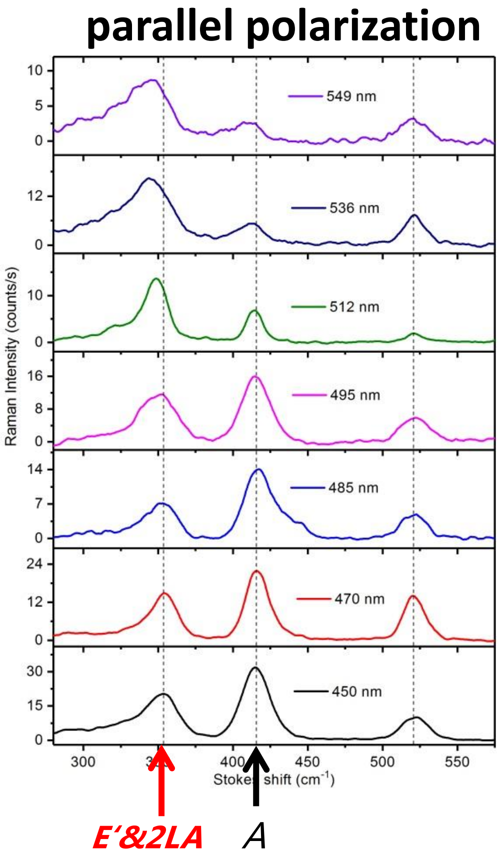


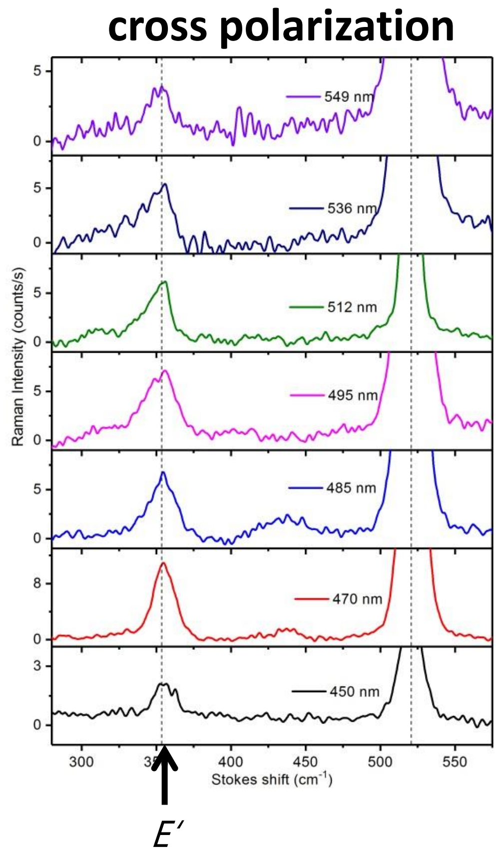
$$I_S = A_S(n+1)$$

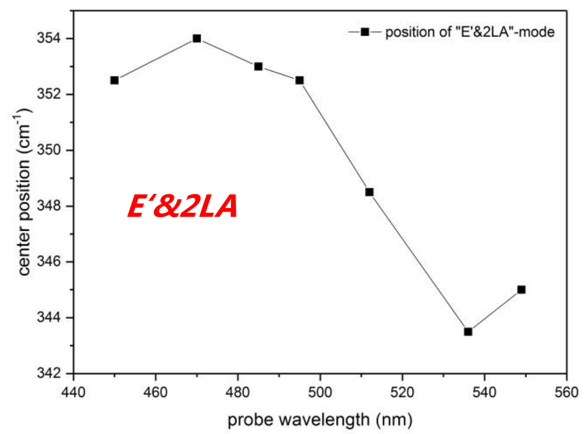
$$\Delta I_S = \cancel{A_S \Delta n} + \Delta A_S$$

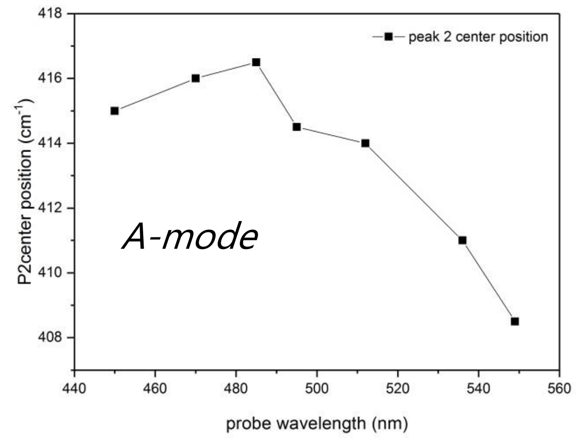
planned measurement: pump/probe with circular polarized light

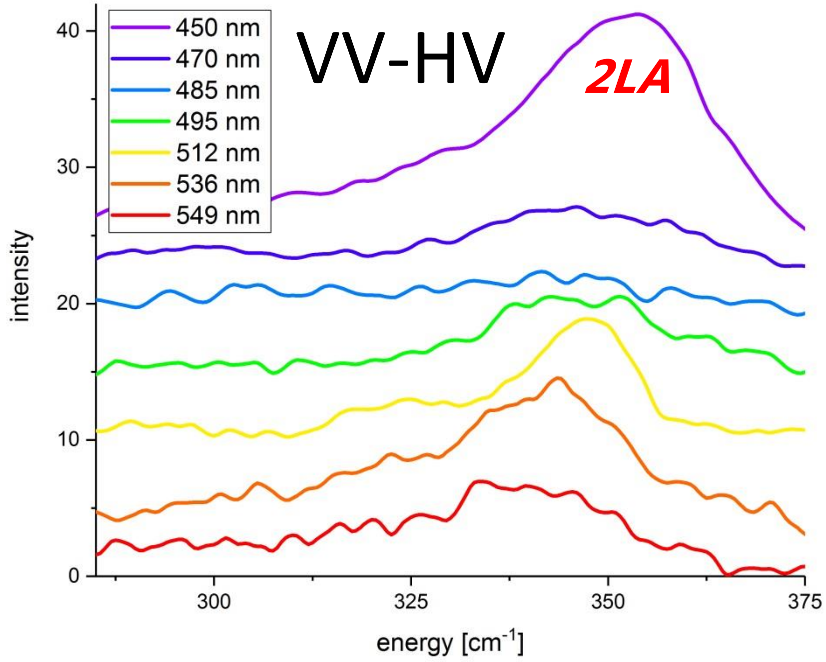
WS2 energy dependent raman

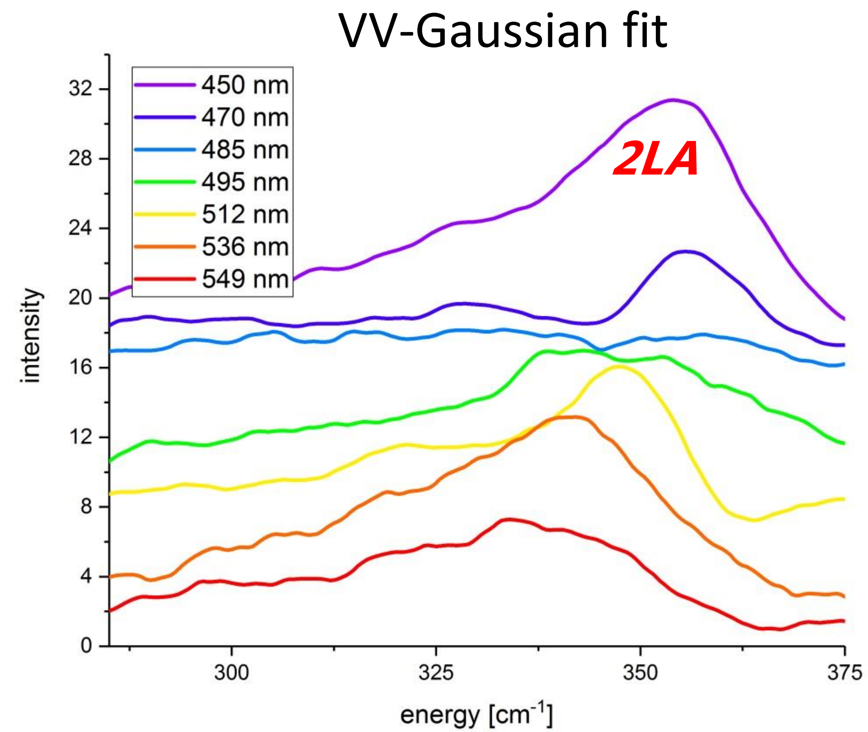


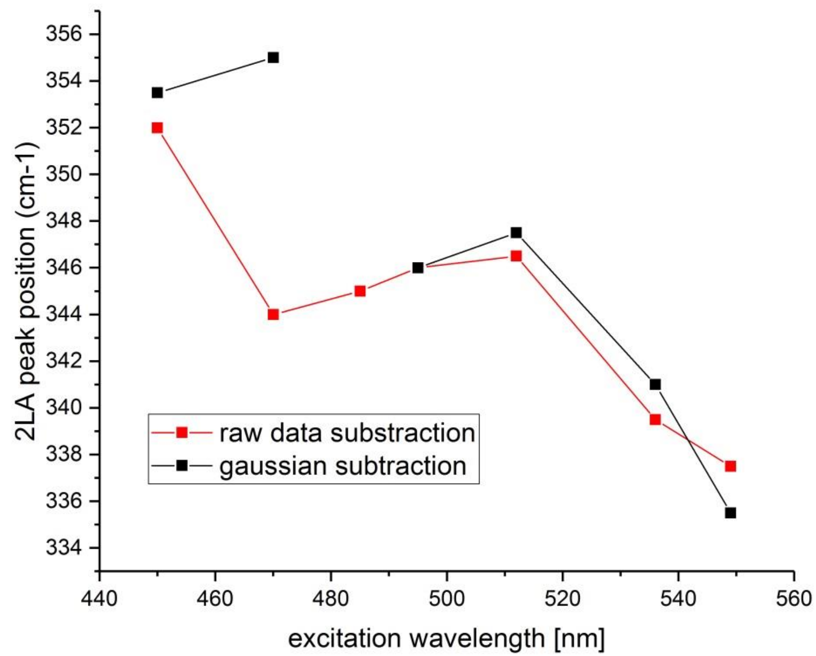


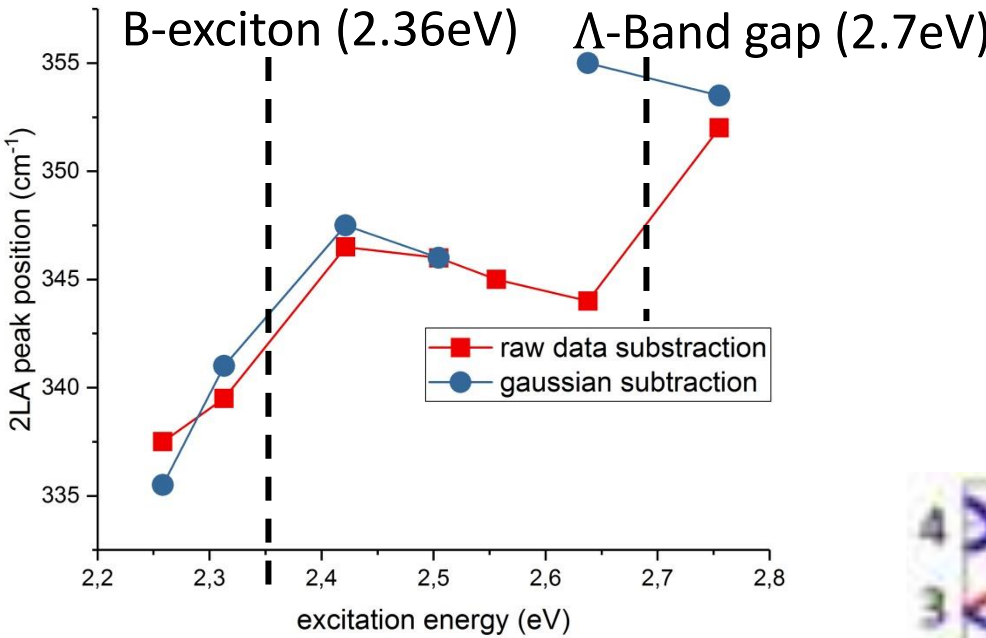


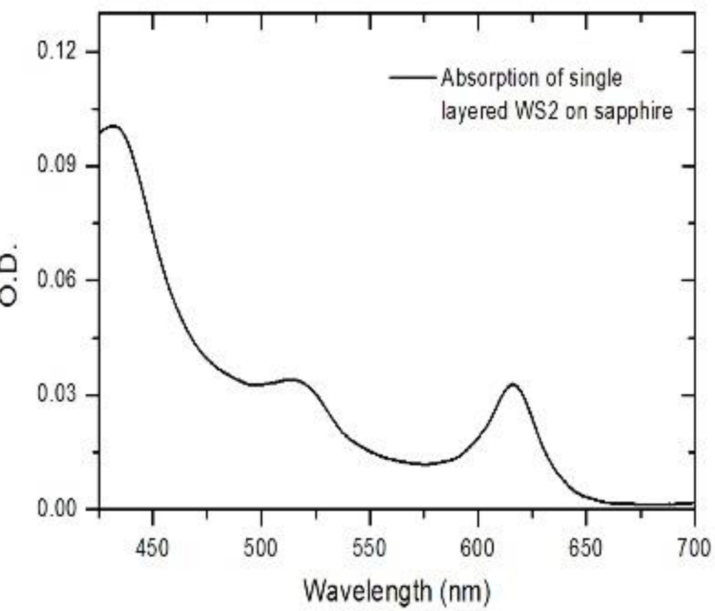



VV-HV
2LA
450 nm
470 nm
485 nm
495 nm
512 nm
536 nm
549 nm
intensity
energy [cm-1]

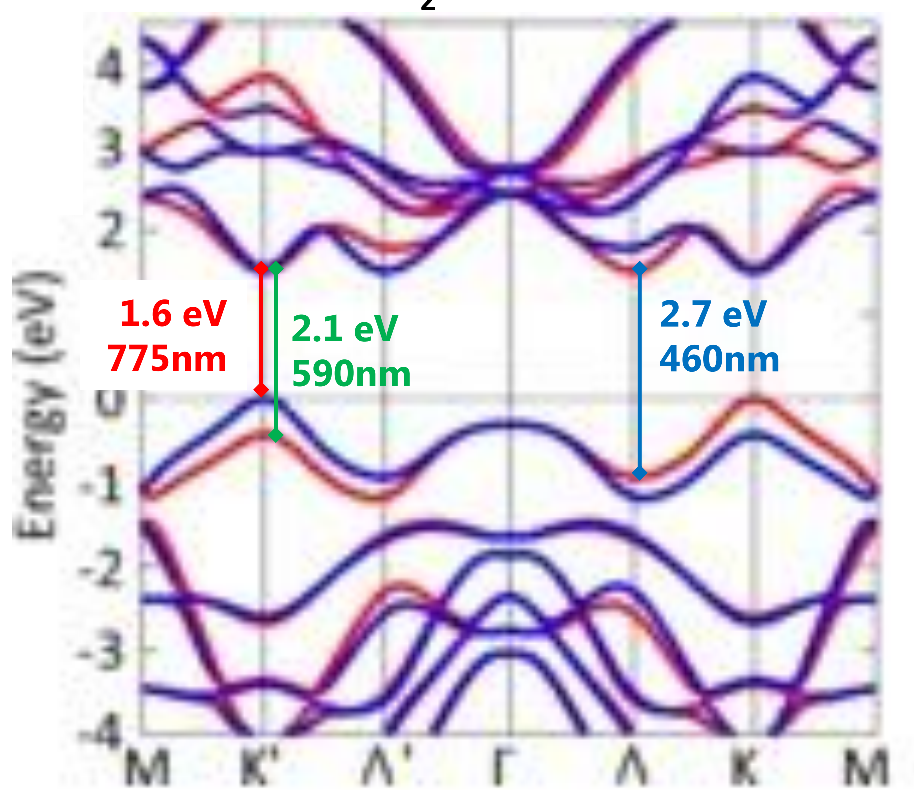

VV-Gaussian fit
2LA
450 nm
470 nm
485 nm
495 nm
512 nm
536 nm
549 nm
intensity
energy [cm-1]


raw data substraction
gaussian subtraction
2LA peak position (cm-1)
excitation wavelength [nm]





WS_2 band structure



Laser energy dependence of valley polarization in transition-metal dichalcogenides

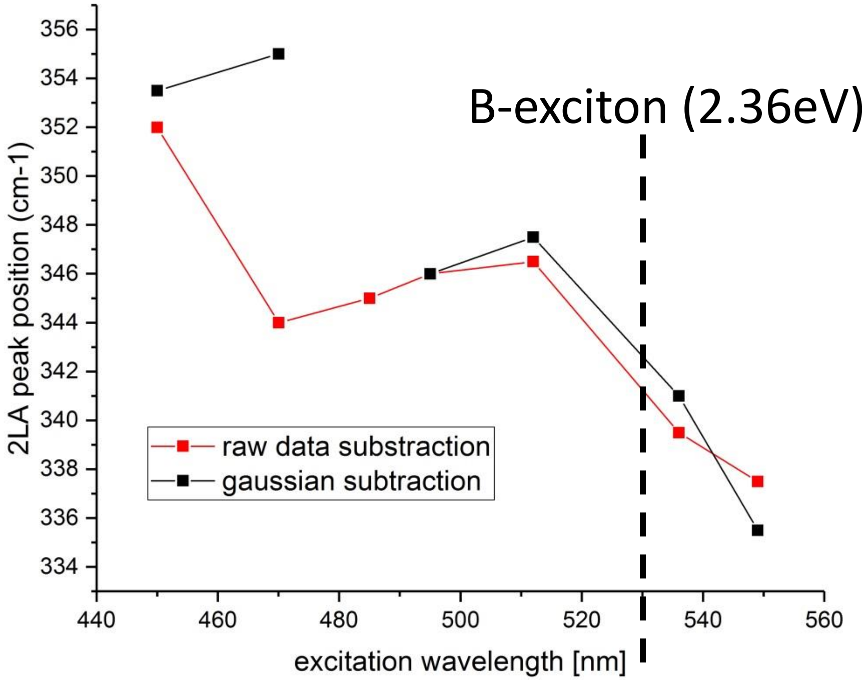
Reasons for dispersive Raman modes:

- double resonant Raman (✓)
- Carries surface depletion, i.e. different wavelength have different penetration depth X
- Photoselective Raman Scattering in inhomogeneous materials X
- Exciton-phonon-coupling ?

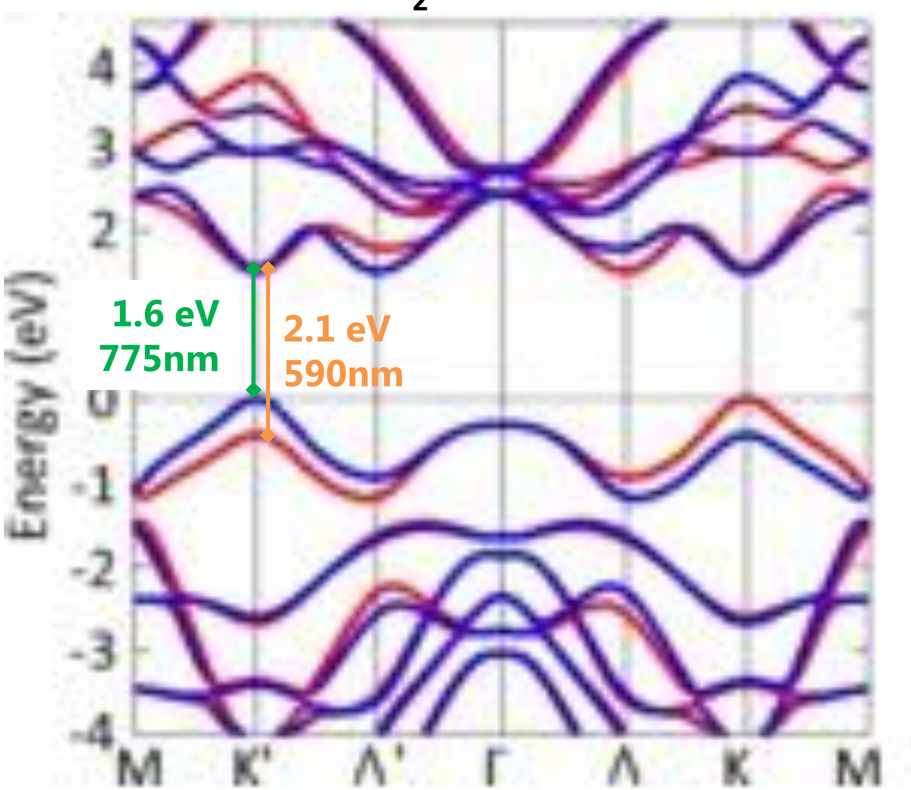
Future experiments

- Use ‘‘cross“-circular polarized light to filter E‘-mode (WS_2)
- Temperature dependence of 2LA-mode (WS_2)
- Model Raman spectrum for WS_2
- Pump and probe with circular polarization (MoS_2 and WS_2)
- Polarization grating on $MoSe_2$ (→ Henning Kuhn, room 338, 16:00, 13.06.)

Thank you!



WS_2 band structure



Laser energy dependence of valley polarization in transition-metal dichalcogenides