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國立臺灣師範大學

透過光激發探究層狀二硫化鉬的激子-聲子交互作用

Exploring Exciton-Phonon Interaction of Layered MoS₂ by Photoexcitation

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研究重點

Polarized Raman spectroscopy can be used to observe the interactions between electron/exciton and phonons when intervalley scattering happens in two-dimensional (2D) materials. Molybdenum disulfide (MoS₂) exhibits double resonance Raman scattering process when the excitation energy is close to the bandgap of the transition metal dichalcogenide (TMD) semiconductor, showing additional Raman peaks in the Raman spectrum. The electron/exciton and phonon interactions in the intervalley scattering processes are mostly determined by deformation potential (DP) and Fröhlich interaction (FI), which are not yet fully understood for the additional Raman peak of b mode observed in MoS₂. In this work, we utilized linearly and circularly polarized light to investigate the polarized phonons in layered MoS₂ under resonant excitation condition. A competition between the FI and DP changes the polarization of b-mode to an opposite polarization state when decreasing the temperature of measurement. These results provide an important technique to investigate light-matter interaction in 2D materials for further developments of their spintronic applications.

研究結果

Second-Order Raman Spectra of MoS₂

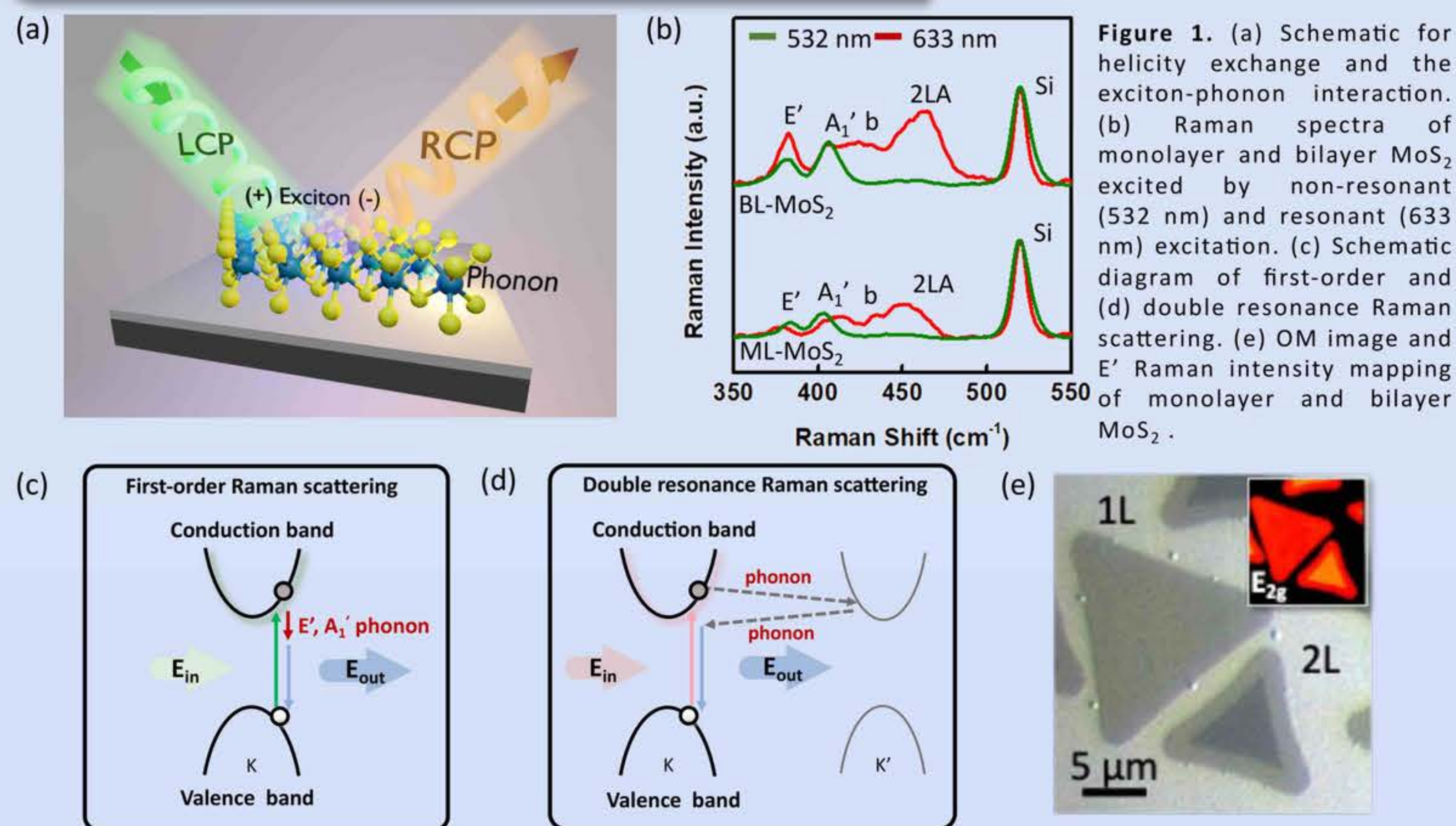


Figure 1. (a) Schematic for helicity exchange and the exciton-phonon interaction. (b) Raman spectra of monolayer and bilayer MoS₂ excited by non-resonant (532 nm) and resonant (633 nm) excitation. (c) Schematic diagram of first-order and double resonance Raman scattering. (d) Energy band diagram showing phonon interactions. (e) OM image of MoS₂ flakes.

Temperature-Dependent Polarized Raman b mode

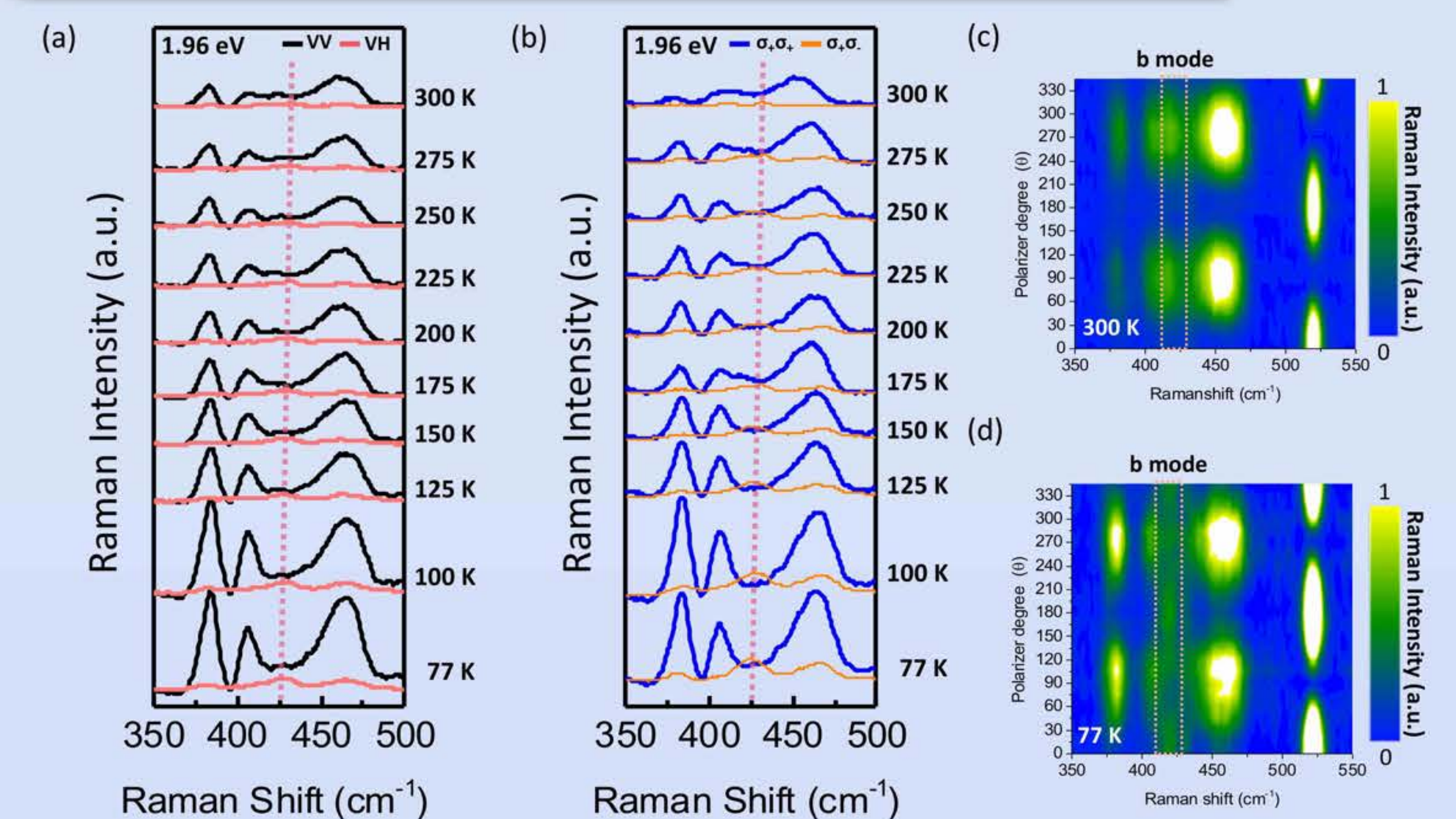


Figure 4. Temperature-dependence of polarized Raman in bilayer MoS₂ excited by (a) linear and (b) circular polarized resonant excitation. (c) Polarized Raman intensity mapping in bilayer MoS₂ measured at 300 K and (d) 77 K under circular polarized excitation.

Experimental Setup

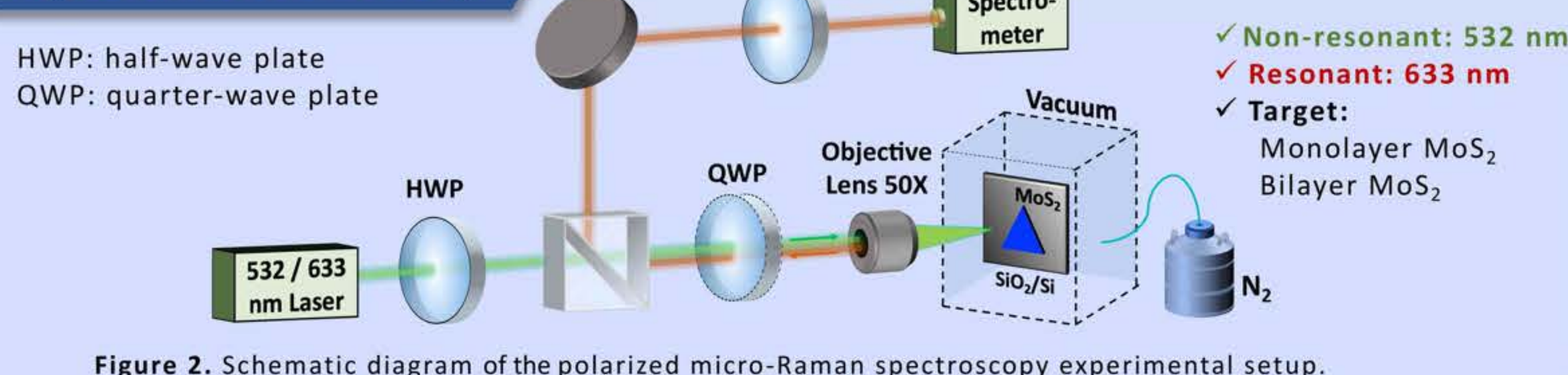


Figure 2. Schematic diagram of the polarized micro-Raman spectroscopy experimental setup.

Degree of Circular Polarization (DoCP) of b mode

$$DoCP (\%) = \frac{I_{\sigma_+ \sigma_+} - I_{\sigma_+ \sigma_-}}{I_{\sigma_+ \sigma_+} + I_{\sigma_+ \sigma_-}} \times 100\%$$

DoCP (%): + : preserved
- : exchanged

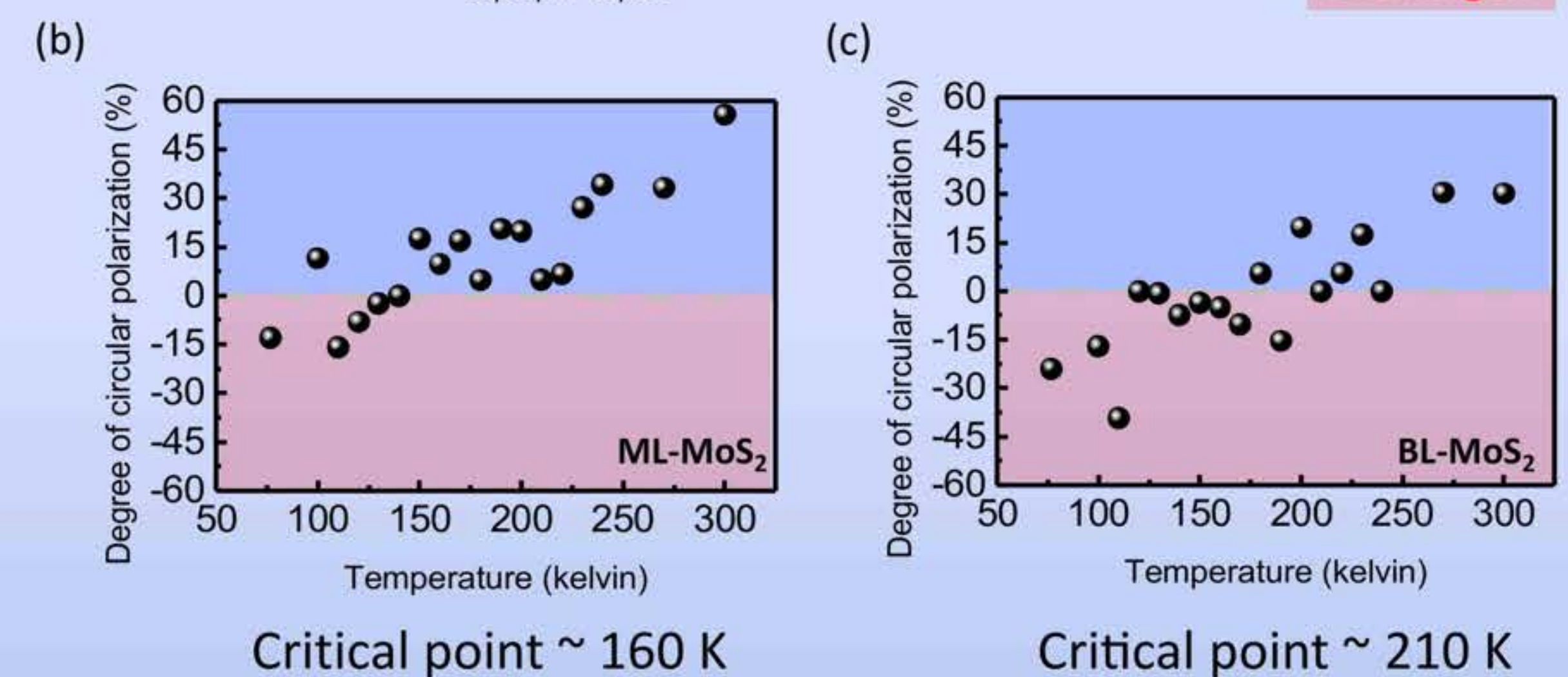


Figure 5. (a) Degree of circular polarization (DoCP) equation. (b) temperature-dependence of DoCP values of b mode in monolayer MoS₂ and (c) bilayer MoS₂.

Polarization-Resolved of MoS₂ Phonon

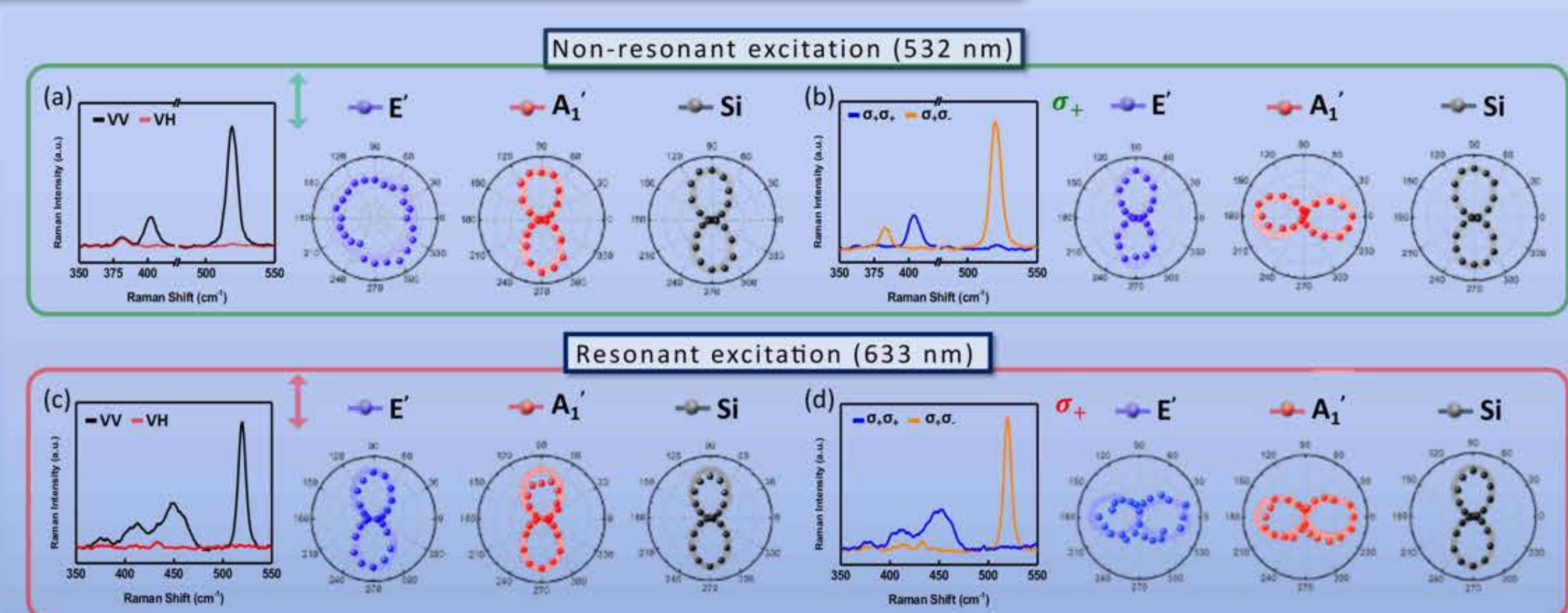


Figure 3. Polarization-resolved Raman spectra and Raman intensity polar plots of E', A₁', and Si in monolayer MoS₂ excited by (a) (b) non-resonant and (c) (d) resonant excitation. The solid curves in the polar plots are numerical results based on Jones calculus and Raman tensors.

Summary & 研究心得

The temperature-dependent behavior of Raman active phonon modes of MoS₂ under double resonance condition has been successfully recorded in this work. Most of the peaks reveal the helicity-preserved polarization state except the b mode. The b mode changes the polarization state from helicity-preserved to helicity-exchanged when decreasing the temperature. The electron/exciton phonon interactions are governed by a fare competition between the FI and DP of the crystal lattice of MoS₂, which is sensitively dependent on the temperature of measurements. The observed b-mode in MoS₂, constituted by LA and TA phonons at K point, is worth to analyze by polarized Raman spectroscopy for more understanding about electron/exciton-phonon interactions in similar 2D TMD materials.

在研究的過程中，有時充滿許多困難，但是這一切的挑戰都很有趣，讓我也很享受自我成長的過程。我特別感謝陸亭樺老師在碩博期間的悉心指導，使我認真投入研究和學習。同樣感謝實驗室的學弟妹和系上助教的協助，讓我在研究歷程中充滿歡樂。最後，由衷感謝家人的支持，讓我能樂觀面對各種挑戰。也謝謝中技社每年都在台博士班學生提供獎助學金的機會，也給我表達謝意的機會。



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