

## Single-beam second-harmonic spectroscopy of valley polarization in molybdenum diselenide

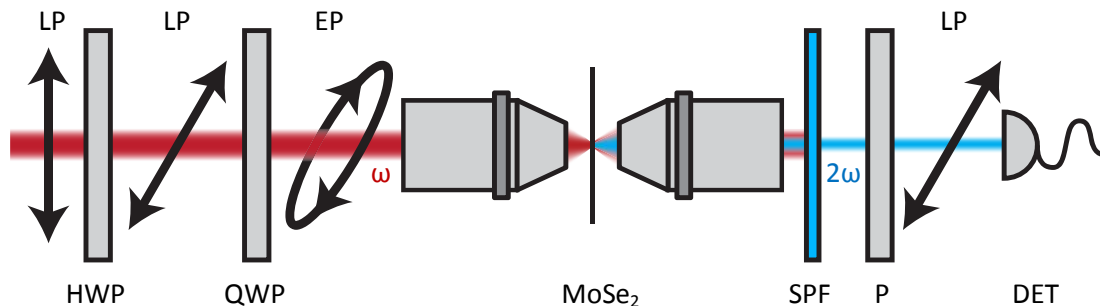
Valleys of a semiconductor are the local minima (maxima) of its electronic conduction (valence) band in the first Brillouin zone. In valleytronics, electron or hole population in each valley is tuned such that when one valley is populated more than the other, valley polarization (VP) is achieved [1-3], in an analogous situation to spin polarization in spintronics. VP can be realized by optically pumping with circularly polarized (CP) light, as demonstrated by Zeng *et al.* [4] and Mak *et al.* [5]. To measure the degree of VP, anomalous Hall effect [2,3] and polarized photoluminescence [4,5] have been exploited.

In this work, we propose to use a single beam of elliptically polarized (EP) ultrafast laser with full polarization control to illuminate a monolayer molybdenum diselenide (MoSe<sub>2</sub>) sample to create VP: the CP component of the beam would induce VP, while the linear polarization (LP) component of the same beam is used to probe the degree of VP by measuring the intensity of second-harmonic generation (SHG). This process will be carried out for incident light polarization with various degree of EP light (from LP to CP). Preliminary results carried out in room temperature are illustrated in Figure 2. The angle with maximum SHG signal ( $\alpha_{\max}$ ) is theoretically calculated to be  $\alpha_{\max} = \Lambda$ , where  $\Lambda = \tan^{-1}[(2 \chi_{\text{int}} \chi_{\text{vp}} \cos \varphi) / (\chi_{\text{int}}^2 - \chi_{\text{vp}}^2)] / 6$ ,  $\chi_{\text{int}}$  = intrinsic second-order susceptibility,  $\chi_{\text{vp}}$  = second-order susceptibility due to VP and  $\varphi$  = phase shift between the susceptibilities.

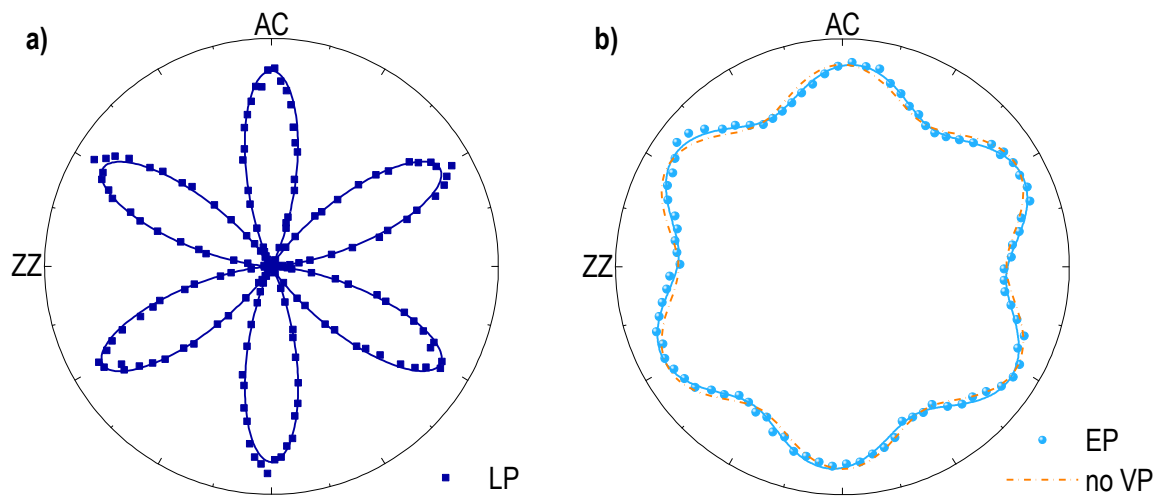
### References

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- [4] Zeng, H. L., Dai, J. F., Yao, W., Xiao, D. & Cui, X. D., *Nature Nanotech.* 8 (2012) 490-493.
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### Figures



**Figure 1:** Experimental setup. Arrows indicate polarization states (LP = linearly polarized, EP = elliptically polarized). HWP = half-waveplate, QWP = quarter-waveplate, SPF = short-pass filter, P = polarizer, DET = detector.



**Figure 2:** Intensity of SHG against MoSe<sub>2</sub> crystal direction (AC = armchair, ZZ = zig-zag) for a) linearly polarized and b) elliptically polarized incident beam ( $\Lambda = 2.7^\circ \pm 0.2^\circ$ ). Orange dash-dotted curve is the expected curve where no valley polarization occurs ( $\Lambda = 0^\circ$ ). Experiments were carried out at room temperature.