Ultrafast control of the valley and spin polarization in Transition-metal dichalcogenides

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Very short valley lifetimes (~10^3-10^6 femtoseconds (fs)) demands the ultrafast control of valley selection on fs time scale. In this work, we explore the possibility to induce valley asymmetry by single-cycle linearly polarized laser pulse in WSe_2 monolayer. By combining the time-dependent density functional theory with the two-band massive Dirac Hamiltonian, we study the various factors that are vital for valley polarization by linearly polarized laser pulse both in linear and non-linear optics limits.

In contrast to long pulses, ultrashort pulses containing few optical cycles, the condition *(maxima of E(t)=0 of A(t))* can be controlled by CEP (φ). φ is the relative phase of the pulse envelope and the oscillating electric field which plays a significant role in the pulse waveform for ultrashort laser pulses. To explore the φ dependence on valley pseudospin, we apply linearly polarized pulses parallel to armchair (Γ-M) and zigzag (Γ-K) directions. Valley polarization does not exist for the field polarized along Γ-M because of the lattice symmetry in that direction. On the other hand, owing to trigonal wrapping, the polarization parallel to Γ-K experiences different band curvature with respect to K and K' point.

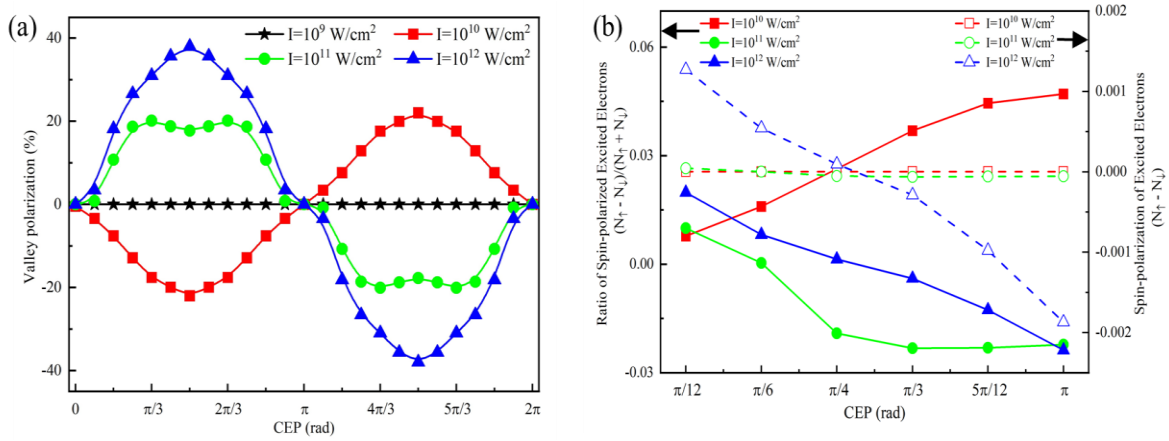

Fig. 1 (a) Valley polarization and (b) Spin polarization of excited charge carriers as a function of CEP.

The valley polarization for weak intensity is zero for all φ confirm the fact that the valley polarization is absent within the linear-optical limit. By increasing the intensity, we enter in the nonlinear regime and the substantial valley polarization is observed. The valley polarization increases gradually with φ and reaches its maximum value at $\varphi = \pi/2$, shows a typical sine wave curve. Valley polarization is robust against field strength but in all cases oscillate as a function of CEP.

The spin polarization ($N_\uparrow - N_\downarrow$) is negligible and independent of intensity and φ. Degree of spin polarization ($N_\uparrow - N_\downarrow$) / ($N_\uparrow + N_\downarrow$) follow the same behavior as valley polarization which shows that spin polarization is also an observable along with valley polarization.