Spin, valley polarization of excited carriers in real materials with time dependent density functional theory

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1. Project Purpose

Two-dimensional (2D) layers provide an excellent platform to investigate and control the valley degree of freedom associated with the energetically degenerate K and K′ valleys, which are formed as the extrema in the Brillouin zone of the valence and conduction bands. This binary valley degree of freedom often designated as the valley pseudospin is readily accessible optically as a consequence of the valley circular dichroism.
The purpose of the project is to manipulate the valley pseudospin in 2D materials. Moreover, we explored the interplay and evolution of the charge, spin and valley excitations coupled to the electron/hole spin because of spin-orbit interactions.

2. Results

In first study, we have developed and implemented 2D Maxwell scheme for extremely thin two-dimensional layers and use the linear-response treatment to calculate the transmission and reflection by 2D conductivity in the weak field limit. WSe_2 monolayer is used to validate our method. A comparison of 2D Maxwell and time-dependent Kohn-Sham (TDKS) method with and without spin-orbit coupling (SOC) is made for electron dynamics calculations. 2D Maxwell calculation of excitation energy and electric current coincides accurately with the TDKS method validates the reliability of our method. To get an understanding of valley pseudospin, we calculated the k-resolved electron populations. We observed that the valley degeneracy is lifted and only K or K′ electron is excited by changing the helicity of the laser.
In another study regarding the valleytronics, we have investigated the possibility of realizing valley Hall effects in blistered graphene sheets. The geometric distortion leads to Van Hove singularities which could induce exchange splitting in otherwise paramagnetic graphene. Unlike the ribbon edges, these atomic-scale blisters are fully immersed within the graphene sheet, hence, its magnetic state is protected from contamination and reconstruction effects that could hamper experimental detection. We have shown that in the presence of exchange splitting, the graphene blister hosts the spin-filtered quantum valley hall effects in units of half $\pm e^2/h$. We propose that we can

have Ω(K) ~ -Ω(-K), the distinguishable chiral valley pseudospin state even in the presence of magnetism. Our study provides a pathway to realize the spin-filtered and valley contrasting hall effects in the absence of external fields in graphene.

3. Roles of the MCRP and its significance

This study was based on computational approaches to correlate, analyze, and understand physical properties of 2D materials. We have verified our method by running simulations on Oakforest-pack super computer, the computational resources are given by MCRP. The calculation of WSe_2 monolayer is carried out with spin-orbit coupling (SOC) using MCRP. To carried out this research, MCRP plays a significance role because the calculations with SOC requires very high usage of memory. In fact, the calculations with SOC is 5 times higher than conventional calculations. MCRP gives us the opportunity to carry such high computational jobs.

4. Future plan

Our future plan to explore the valley pseudospin by linear polarized laser pulses. Future plan to investigate the valley excitation in 2D WSe_2 monolayer on a few femtosecond timescales. Moreover, the effect of intensity and carrier envelope phase dependence on valley population will be checked.

5. Publications and conference presentations

(1) Journal papers

Farooq, M. U.; Hashmi, A.*; Ono, T.; Huang, L. Spin–Valley Hall Phenomena Driven by Van Hove Singularities in Blistered Graphene. npj Computational Materials 2020, 6, 1–8.

Supercomputer	Use	Allocated resources*	
		Initial resources	Additional resources
Cygnus	No		
Oakforest-PACS	Yes	192,000	
*in units of node-hour product			