Optical selection rule of monolayer transition metal dichalcogenide by an optical vortex

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Abstract. We have derived the optical selection rule of monolayer transition metal dichalcogenide (TMD) for various optical vortices. Monolayer TMD, which is two-dimensional direct semiconductor, has a twofold valley degree of freedom. The valley degree of freedom can be controlled by circular polarized light. However, it is not clear how the orbital angular momentum of light relates to the valley degree of freedom. Here we clarified that the orbital angular momentum of light modifies the optical selection rule at valley points and the selection rule reflects the three-fold rotation symmetry of monolayer TMD. We expect that this modified selection rule broadens the research field of two-dimensional layered materials and spin-valleytronics.

1. Introduction

Since the famous discovery of graphene[1], two-dimensional layered materials have attracted significant interest because of their excellent electronic and physical properties. Monolayer transition metal dichalcogenide (TMD) such as MoS₂ has direct bandgaps at the two corners of the Brillouin zone ± K points (see Fig. 1). Thus, in addition to charge and spin, carriers in monolayer TMD have an extra freedom, which is called “valley degree of freedom”. The valley degree of freedom can be selectively operated using circularly polarized light due to optical selection rule[2]. Furthermore, the valence bands at ± K-points have large valley-contrasting spin splitting (0.1 ~ 0.5eV) due to strong spin-orbit coupling and broken inversion symmetry. Therefore, monolayer TMD is expected to a proper platform for spin-valleytronics[3, 4].

The operation of spin and valley mentioned above uses the spin angular momentum (SAM) of light. Here, the SAM corresponds to circular polarization of light. The SAM of light has been applied in a wide range of fields and we have often considered only SAM as the angular momentum of light. However, light with finite orbital angular momentum (OAM) has attracted much attention in the field of optics[5]. Such a light has spiral equiphasic surface and corkscrew potential (see Fig. 2), so it is often called “optical vortex”. Applications of optical vortex are actively researched, e.g., nano chiral laser ablation[6] and optical manipulation[7]. However, it has not been clear how the OAM of light affects carriers in material.

In this work, we focus on an interaction between the OAM of light and carriers of monolayer TMD. To investigate the interaction, we aim to clarify the optical selection rule for various optical vortices. In general, it is believed that the optical selection rule is changed by the presence of OAM of light[8, 9]. However, the relation between the OAM of light and the valley degree of freedom is still elusive.
Figure 1. A schematic of the band structure of monolayer TMD. The band edges at the corners (±K) of 2D hexagonal Brillouin zone. The inequivalent valleys can be operated by circular polarized light.

Figure 2. A schematic of optical vortex beam. This picture shows phase variation, equiphase surface and intensity distribution about \( l = 0 \) to \( l = 3 \). This beam have an azimuthal phase dependence of the type \( \exp(iℓφ) \). In the case of \( ℓ \neq 0 \), optical vortex has phase singularity and helical equiphase surface. The beam intensity on the propagation axis is zero.

2. Method
We have derived the optical selection rule of monolayer TMD by various optical vortices. Early theoretical studies reveals that the band edges at K and −K are mainly dominated by transition metal d-orbitals in monolayer TMD[10]. The trigonal prismatic coordination classifies the transition metal d-orbitals into three groups with irreducible representations: \( A(d_{z^2}) \), \( E(d_{xy}, d_{x^2-y^2}) \) and \( E'(d_{xz}, d_{yz}) \) of the \( C_{3h} \) point group. The wave functions of conduction and valence band at K and −K are

\[
|\Psi_c⟩ = |d_{z^2}⟩, \quad |\Psi_v⟩ = \frac{1}{\sqrt{2}} \left( |d_{x^2-y^2}⟩ + iτ|d_{xy}⟩ \right),
\]

where the index \( c(v) \) means conduction (valence) band and \( τ = ±1 \) is the valley subscript.

In order to clarify inter-band transition at K and −K, we calculated the optical transition amplitude between the d-orbitals mentioned above. The calculated transition matrix is

\[
⟨Ψ_f|p \cdot A^l + A^l \cdot p|Ψ_i⟩ \sim \frac{2im}{ℏ} (ΔE) ⟨Ψ_f|r \cdot A^l|Ψ_i⟩,
\]
where $|\Psi_i\rangle$ indicates the initial state, $|\Psi_f\rangle$ is the final state, $\Delta E$ means the energy difference between them. Here, we assume that the optical vortex is Laguerre-Gaussian beam. Its vector potential is described as

$$A^l(r, \phi, z) = e_A \sqrt{\frac{2p!}{\pi(p + |l|)!\omega_0}} \left[ \frac{r\sqrt{2}}{\omega_0} \right]^{[l]} L_p^{[l]} \left( \frac{2r^2}{\omega_0^2} \right) \exp \left( -\frac{r^2}{\omega_0^2} \right) \exp (il\phi),$$  

(3)

where $l \in \mathbb{Z}$ characterizes the OAM, $p \in \{0, 1, 2, \cdots \}$ indicates the radial index, $\omega_0$ is the beam waist and $e_A$ is the polarization vector

$$e_A = \begin{cases} (1/\sqrt{2}, i/\sqrt{2}, 0) & \text{(right-handed circular polarization)} \\ (1/\sqrt{2}, -i/\sqrt{2}, 0) & \text{(left-handed circular polarization)} \end{cases}$$  

(4)

Here, we have considered only the case for $p = 0$ because the radial component of the beam does not affect the selection rule. In this calculation, we have not considered the spatial spread of the d-orbital wave function. The ratio of the spread and the beam waist $\omega_0$ affects the optical transition intensity. However, the selection rule itself does not depend on the ratio.

We calculated the transition matrix mentioned for the case of $l = -4$ to $l = +4$. When the transition matrix is zero, the transition is forbidden. When the matrix is finite, the transition is allowed. In calculating the transition matrix, we took into account the three-fold rotation symmetry of monolayer TMD.

3. Result

The resultant selection rules are summarized in the Table 1 and Fig. 3. These table and figure indicate the transition from the valence band to the conduction band at $\pm K$. For the circularly polarized plane wave ($l = 0$), the selection rule is the well-known one[2]. One can see that the OAM of light modifies the optical selection rule. Furthermore, we clarified that the selection rules are the same for every 3 of $l$, e.g., the selection rules coincide for the cases of $l = 0$ and $l = \pm 3$. This result reflects the three-fold rotation symmetry of monolayer TMD.

Table 1. Optical selection rule at $\pm K$. $l$ characterizes the orbital angular momentum of light ($l = -4 \sim +4$). RCP and LCP mean right-handed circular polarization and left-handed circular polarization respectively. Check mark indicates that the corresponding transition is allowed, and cross mark indicates that the transition is forbidden.

| $l$ | -4 | -3 | -2 | -1 | 0  | +1 | +2 | +3 | +4 |
|-----|----|----|----|----|----|----|----|----|----|
| $K$ | RCP| × | × | ✔ | × | × | ✔ | × | ✔ |
|     | LCP| × | ✔ | × | × | ✔ | × | × | × |
| $-K$| RCP| × | ✔ | × | × | ✔ | × | × | ✔ |
|     | LCP| ✔ | × | × | ✔ | × | × | ✔ | × |

4. Summary

We have investigated selection rules of monolayer TMD for various optical vortices. We clarified that OAM of light modifies the optical selection rule at $\pm K$ points and the result reflects the three-fold rotation symmetry of monolayer TMD. The modified selection rules have the potential
of new physics and application. This achievement broadens the research fields of two-dimensional layered materials and spin-valley physics.

It is considered that the optical vortex radiation to semiconductors causes new light-matter interactions. In future, we will reveal an interaction between the OAM of light and the carrier spin of monolayer TMDs. We expect that the new interaction makes a nontrivial spin-texture of the itinerant electrons, like skyrmion[11], without strong electron-electron correlations. Furthermore, the spin-texture may reflect the valley information because of strong spin-valley coupling. Finally, we want to create the new field of spin-valleytronics by optical vortex.

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Reference

[1] Novoselov K S, Geim A K, Morozov S V, Jiang D, Zhang Y, Dubonos S V, Grigorieva I V and Firsov A A 2004 Science 306 666
[2] Cao T, Wang G, Han W, Ye H, Zhu C, Shi J, Niu Q, Tan P, Wang E, Liu B and Feng J 2012 Nat. Commun. 3 887
[3] Xiao D, Liu G B, Feng W, Xu X and Yao W 2012 Phys. Rev. Lett. 108 196802
[4] Xiaoqiong X, Wang Y, Di X and Tony F. H 2014 Nature Physics. 10, 343-350
[5] Xiaodong X, Wang Y, Di X and Tony F. H 2014 Nature Physics. 10, 343-350
[6] Allen L, Beijersbergen M W, Spreeuw R J C, and Woerdman J P 1992 Phys. Rev. A. 45, 8185
[7] Toyoda K, Takahashi F, Takizawa S, Tokizane Y, Miyamoto K, Morita R and Omatsu T 2013 Phys. Rev. Lett. 110 143603
[8] Garces-Chavez V, McGlone D, Padgett M J, Dultz W, Schmitzer H and Dholakia K 2003 Phys. Rev. Lett 91 093602
[9] Picon A, Benseny A, Mompart J, Vazquez de Aldana J R, Plaja L, Calvo G F. and Roso L 2010 New Journal of Physics 12, 083053
[10] Schmieglow C T, Schulz J, Kaufmann H, Ruster T, Poschinger U G and Schmidt-Kaler F 2016 Nat. Commun. 7 12998
[11] Coelhoorn R, Haas C and de Groot R A 1987 Phys. Rev. B 35, 6203
[12] Yu X Z, Onoue Y, Kanazawa N, Park J H, Han J H, Matsui Y, Nagaosa N. and Tokura Y 2010 Nature 465, 901-904