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Spin-valley physics in 2D crystals

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Abstract

Transition metal dichalcogenide (TMD) is attracting growing interests as two dimensional (2D) crystals beyond graphene. In particular, the absence of inversion symmetry and strong spin-orbit interaction in TMD based 2D crystals offer novel spin-valley functionalities, making TMD an extremely unique system. Here we report the direct observation of the out-of-plane spin polarization which is strongly coupled with valley degree of freedom. Also, we realized WSe_2 based light emitting transistor, which allows us an electrical control of the circular polarization.

1. Introduction

The crystal structure of monolayer transition metal dichalcogenide (TMD) MX_2 (M = Mo and W, X =S, Se, Te) has a honeycomb lattice structure with broken inversion symmetry, as shown in the top panel of Fig. 1. In terms of electronic structure, the monolayer MX_2 is known to have a direct band gap of 1 ~2 eV at the corners of hexagonal Brillouin zone called *K* and *K'* points (right panel of Fig. 1) [1], in contrast to the bulk crystal which has an indirect band gap. Thus the monolayer TMD is regarded as a gapped graphene and shows a high ON-OFF ratio in the field effect transistor (FET) operation. The bands at *K* and *K'* points, which are called valley, are energetically degenerate. If one is able to intentionally control the occupation of these bands by electron, valley can be regarded as a pseudo-spin, a new degree of freedom in solids. The manipulation of valley is called valleytronics and, in fact, the selected excitation of valleys has been realized by shining the circular polarized light [2,3] . Another important feature of TMD is the large spin-orbit interaction (SOI), which causes a spin-splitting in the top of the valence band as shown in the right panel of Fig. 1. The spin splitting is theoretically predicted to be 0.1 eV and 0.4 eV in Mo and W

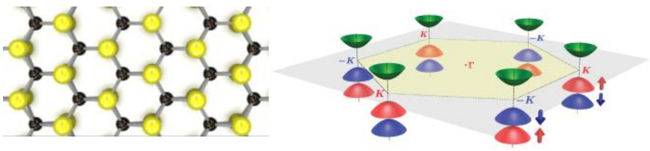

Fig. 1 Crystal structure (left) and band structure (right) of monolayer TMD..

compounds, respectively [4]. The spin polarization is out-of-plane, and is strongly coupled with the valley degree of freedom. As seen in the right of Fig. 1, the *K* and *K'* valley spins are oppositely polarized, and consequently the total spin polarization is cancelled.

2. SARPES observation of valley dependent spin polarization

Although the spin-split band structure is a widely accepted model, the experimental confirmation of the valley dependent spin polarization has not been made thus far, because the spin- and angle-resolved photoemission spectroscopy (SARPES) experiments are the only way to prove this, but the SAPES requires a sample surface as large as around 1 x 1 mm^2. However, the size of monolayer TMDs is typically 10 x 10 μm^2, when they are fabricated by mechanical exfoliation. On the other hands, once we choose single crystals with sufficient sizes, it usually takes the 2H polytype, where the layers are stacked so that the space inversion symmetry of the monolayer is restored.

To overcome this difficulty, we took advantage of rich polytypes of TMDs. For the case of MoS_2, in addition to the most stable 2H phase, the 3R phase is known to exist as a metastable phase. The space group of the 3R structure is *R3m*, and its unit cell is composed of a trilayer stacked in such a way that the inversion symmetry is kept broken in the bulk form. In this noncentrosymmetric crystal structure, the intrinsic properties of TMD monolayers including the valley dependent spin polarizations are supposed to be maintained. We successfully grew single crystals of the 3R-phase, and confirmed by means of ARPES experiments that the in-plane electronic structures are almost identical to those of the 2H-phase, reflecting the strongly 2D nature of both forms of MoS_2. Figure 2 displays the spin dependent band dispersion of the valence band top at the *K* point. The top of the valence band are up-spin polarized, whereas the second top band are down-spin polarized. The successfully observed the out-of-plane spin polarization in the valence band at the *K* and *K'* points, which displays fair agreement with the first principles band calculations taking the SOI into account. This is the first proof of the valley dependent spin polarization expected in the monolayer MoS_2, by utilizing the bulk single crystals with noncentrosymmetric layer stacking pattern of the 3R-form [5].

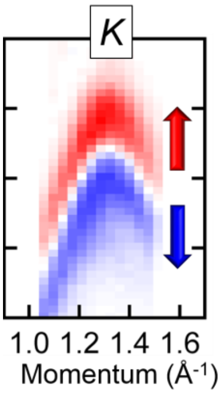

Fig. 2 Spin dependent band dispersion at the K point in 3R-MoS_2. Red and blue colors indicates the spin polarization.

3. Electrically tunable chiral light source

Because the electronic states of the two valleys have different chiralities due to the inversion asymmetric crystal structure, interband transitions at K and K' points are featured with right- and left-handed circular polarized light (σ_+ and σ_-), respectively (valley circular dichroism). These features are suppressed in bulk single crystals because of the appearance of the inversion symmetry associated with the variation of the band structure from direct gap to indirect gap. Circular polarized photoluminescence from TMDs has been experimentally observed for monolayers [2, 3] or electrically biased bilayers pumped with circular polarized incident light [6]. In the following, we show current induced circular polarized electroluminescence (EL) from *p-i-n* junctions in monolayer and multilayer WSe_2 [7].

TMD FETs are well known for their ambipolar operation, where both electrons and holes contribute to channel current dependent on the gate voltage applied [8, 9]. Fig. 3 displays I_{DS} –V_{DS} relations of the WSe_2 electric doubly layer transistor (EDLT), which is a kind of FETs using ionic liquids as gate dielectrics.

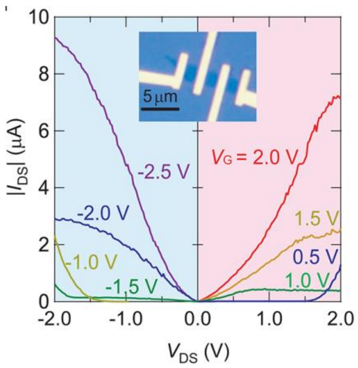

Fig. 3 Output curve (I_{DS} vs. V_{DS}) of WSe_2 EDLT. (Inset) Typical optical micrograph of the device.

Using this ambipolar transistor, we are able to fabricate electric-field-induced *p-i-n* junction in the WSe_2 channel as shown in Fig 4. In the ambipolar transistor, the current flows in a forward direction, resulting in electroluminescence (EL). A typical EL spectrum of the monolayer WSe_2-EDLT is shown in Fig. 4, showing clear circular dichroism. This is quite unexpected, because we are not injecting neither valley or spin polarized current into the device, although the observation of circular polarized PL indicates there exists some mechanism which generates valley polarization. Another interesting feature we encounter is that the circular polarization can be changed by reversing the direction of current [7].

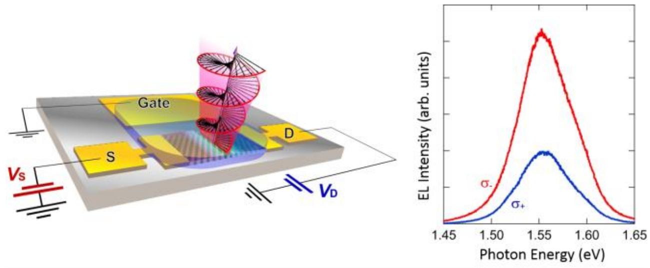

Fig. 4 Schematic of ambipolar light emitting transistor of WSe_2 EDLT (right). Circular polarized EL spectrum (left).Typical optical micrograph of the device.

This prominent feature is possibly understood by taking the trigonal warping of the valence band into account. The injected current itself is not basically valley polarized, however the recombination rate at the p-i-n junction may be different between K and K' points, due to the trigonal warping of the valence band.

Since the circularly polarized light is highly useful in various applications, there is a strong demand for a light source endowed with both the compactness for high integration and the electrical controllability of the polarization. The present light emitting transistor of TMD might be a possible model for the electrically switchable chiral light source based on the valleytronic functionalities.

Acknowledgements

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