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Room Temperature Valley Polarized Light-Emitting Diodes of Monolayer Transition Metal Dichalcogenides

Owing to broken spatial inversion symmetry and spin-orbit interaction in monolayer transition metal dichalcogenides (TMDCs), their electronic properties at the band edge are associated with the two inequivalent (K and -K) valleys, resulting in the interband transition coupled with optical helicity [1,2]. Indeed, the valley polarization have been exclusively controlled by circularly polarized light pumping [2]. These features motivate us to develop valley-polarized light-emitting diodes (LEDs) that can electrically generate circularly polarized light emission. Although several researches have reported chiral electroluminescence (EL) from TMDCs, the solid manner for electrical control valley polarized EL have remained unclear [3,4]. This lead to the limited utility of relevant valley polarized LEDs, for instance, circularly polarized EL have mostly observed at low temperature (< 80 K). Therefore, establishing the approach to produce valley polarized LEDs operated at room temperature is highly required for the development of TMDC-based valley functional device applications.

To understand operational mechanism for circularly polarized EL, the detailed evaluations in terms of electric field dependence, temperature dependence, and importantly, spatial imaging of polarized light emission should be addressed. However, current methods for fabricating TMDC LEDs have adopted complicated device configurations such as transistors and heterostructures using tiny exfoliated samples, and thus, this fundamental barrier has made investigating circularly polarized EL properties of TMDCs inevitably difficult [3,4]. Here, we newly propose a versatile and simple approach to generate light emission in TMDCs [5,6]. The proposed device only needs two electrodes deposited onto TMDCs, followed by spin-coating ion gels, a mixture of ionic liquid and triblock co-polymer (Fig. 1). We apply this method to chemical vapor deposition (CVD)-grown single-crystalline WSe_2 and WS_2 monolayers to achieve polarized EL imaging and spectroscopy.

Figures 2a and 2b shows optical micrograph and EL image obtained in WS_2 LEDs near room temperature (280 K). We observed clear light emission between two electrodes, resulting in EL generation due to electrolyte-induced p-i-n junctions (Fig.1). Owing to direct EL observations, we performed spatial polarization-resolved EL spectroscopy. Figures 2c and 2d exhibit polarization-resolved EL spectra obtained at two different positions of WS_2 flake, in which each spectrum was recorded inside crystal and crystal edge regions, respectively. Only small EL polarization was obtained at lower temperature (< 40 K) inside crystal regions (Fig. 2d). In contrast, larger EL polarization was observed at higher temperature (> 100 K) at crystal edge regions (Fig. 2c). Most importantly, this large EL polarization was robustly remained up to 280 K. These results suggest the position-dependent distinct EL polarization mechanism in TMDCs. In order to examine the origin of robust circularly polarized EL, furthermore, we compare these EL results with photoluminescence mapping done in same crystal. As a result, we found out that the local strain induced at crystal edge regions due to lattice mismatch would play a significant role to create robust EL polarization in TMDCs. Our observations provide possible ability to construct practical TMDC-based atomically thin chiral light sources for future opto-valleytronic applications.

References

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Figures

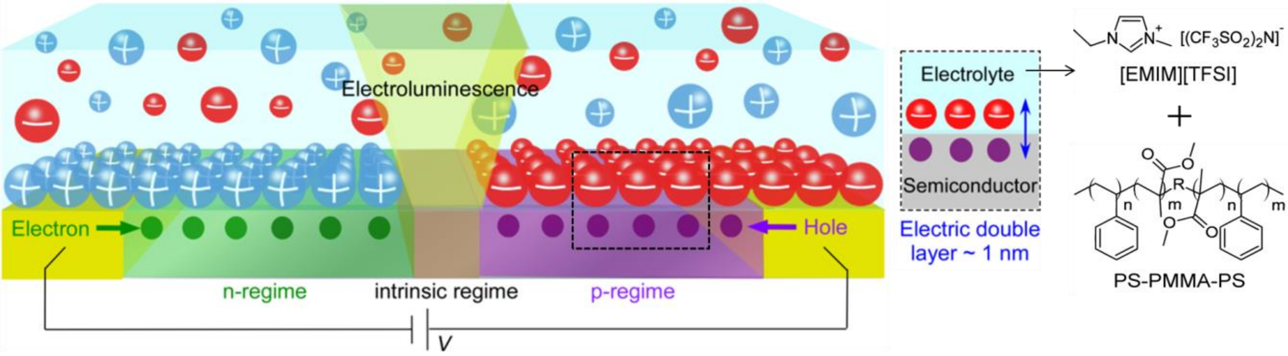

Figure 1: Schematic illustrations of a proposed light-emitting device, an ionic liquid, and a triblock copolymer.

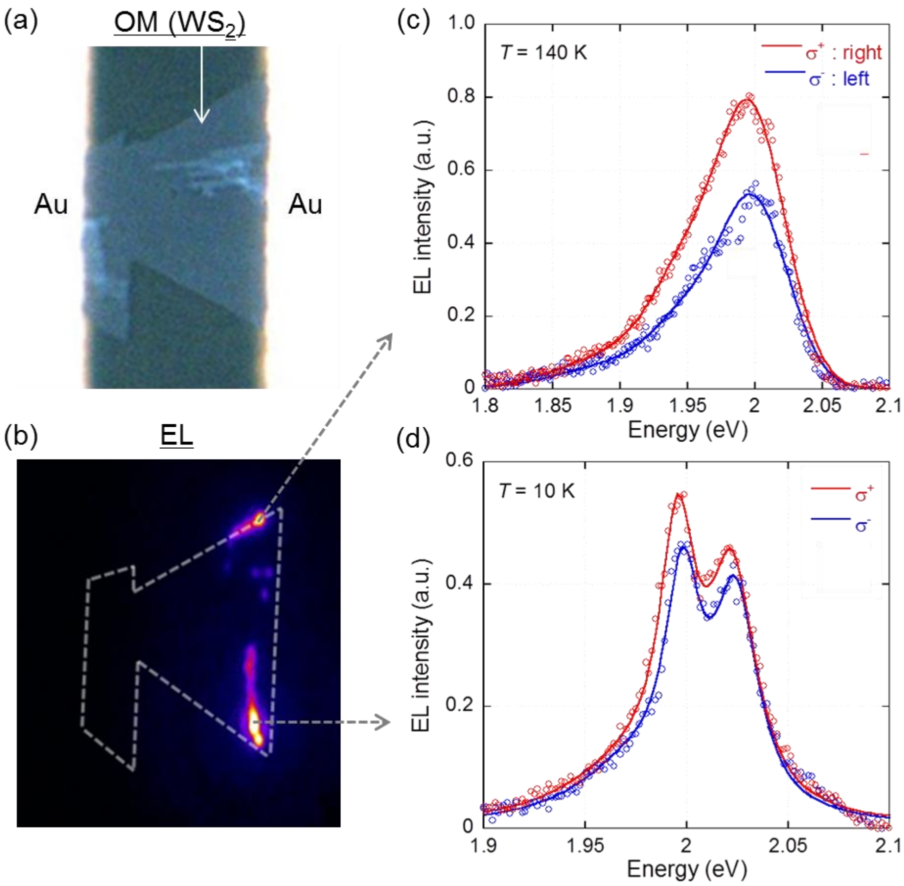

Figure 2: (a) Optical micrograph and (b) EL image of a CVD-grown single-crystalline monolayer WS_2 light-emitting diode. Polarization-resolved EL spectra measured at temperature of (c) 140 K and (d) 10 K, in which each spectrum is recorded at different positions of a WS_2 flake.