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Direct Electroluminescence Imaging of Transition Metal Dichalcogenides

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Abstract

We newly propose a versatile electrolyte-based device structure for fabricating light-emitting device using transition metal dichalcogenide (TMDC) monolayers. Our approach enables direct electroluminescence (EL) imaging and spectroscopy to investigate unique optical properties of TMDCs, and thus, firstly reveals robust circularly polarized light emission near room temperature. This result raises the practical utility of monolayer TMDCs for designing chiral light-emitting devices.

1. Introduction

Emerging direct bandgap and unique electronic structure, arising from broken spatial inversion symmetry, in monolayer transition metal dichalcogenides (TMDCs) provides a new platform for exploring novel optoelectronic functionalities and devices [1-3]. One of the most promising device applications is the light-emitting diodes (LEDs). In general, LEDs require intentional doping techniques, such as ion implantation and chemical doping, to form p-n junction, however, reliable doping methods for monolayer TMDCs have not yet been fully established. Therefore, the fabrication of TMDC LEDs has been limited by using complicated device configurations, and this fundamental barrier has made investigating electroluminescence (EL) properties of TMDCs inevitably difficult [4-6]. To overcome this issue, we recently established electrolyte-based doping methods [7-9]. Here, we newly propose a versatile and simple approach to generate light emission in TMDCs [10,11]. We apply this method to chemical vapor deposition (CVD)-grown polycrystalline and single-crystalline WS_2 and WSe_2 monolayers to achieve direct EL imaging and spectroscopy. Our demonstrated EL imaging is advantageous to evaluate polarized EL properties originating from topological features of TMDCs, and thus, we firstly reveal robust circularly polarized EL near room temperature.

2. Experiment

Device fabrication

The monolayer TMDC LEDs was fabricated only by two steps. First, two golds electrodes were thermally deposited onto CVD-grown polycrystalline and single-crystalline WSe_2 and WS_2 monolayers [12]. And then, ion gels [13], a mixture of ionic liquid, [EMIM][TFSI], and triblock co-polymer, PS-PMMA-PS, were spin-coated on TMDC surface to complete fabrication process (Fig. 1 top). It is worth noting that, because the thickness of ion gel films is less than 100 nm, it is enough transparent to perform spectroscopic measurements. As shown in the schematic of Figure 1, just by applying voltage between two electrodes, ion redistributions induce both electron and hole accumulations originating from electric double layers formed on TMDC surfaces. This dynamic electrostatic doping process finally leads to p-i-n junction formation to generate EL.

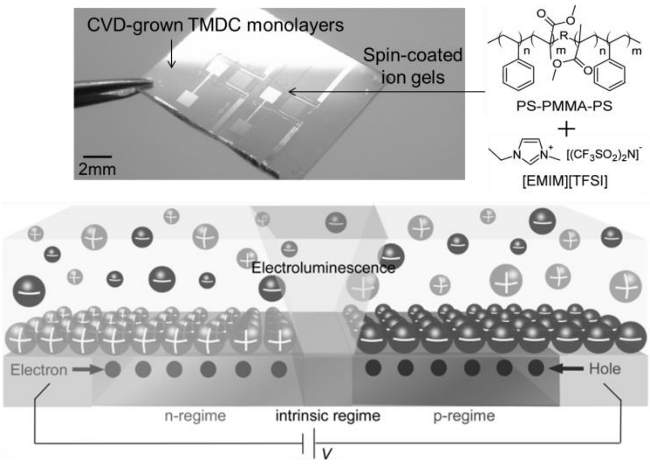

Fig. 1 A proposed electrolyte-based light-emitting device.

Measurement

For the EL characterizations, the devices were set inside cryostat with vacuum condition under 10^{-5} Torr, which can control stage temperature from 300 K to 5 K. The electrical measurements were controlled by source meter and electrometer. The EL imaging and spectroscopy were simultaneously collected through objective, in which the emitted lights were separated by half mirror, one optical path is getting in a CCD camera, and the other is for spectrometer. In addition, the polarization-resolved EL measurements were performed by optical set-up, where the emitted lights were passed through a 1/4 wave plate and separated into two orthogonally polarized lights. The separated light was depolarized followed by focusing to two spots on a CCD camera, in which two beam spots correspond to right- (σ^+) and left- (σ^-) handed circularly polarized light, respectively.

Results and Discussion

Figure 2 shows optical micrograph and EL image for both polycrystalline and single-crystalline WSe_2 monolayers measured at room temperature. We observed clear light emission between two electrodes, resulting in EL generation due to electrolyte-induced p-i-n junctions. Importantly, we expanded this light-emitting structure to various TMDCs, such as MoS_2, $MoSe_2$, WS_2, ReS_2, and MoS_2-WSe_2 lateral heterojunctions, and successfully observed light emission, which directly indicates the versatility of our proposed approach for building TMDC light-emitting devices [14].

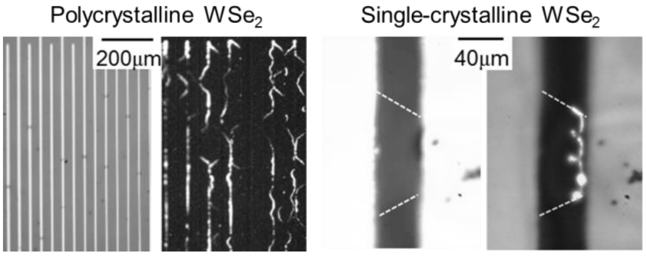

Fig.2 Optical micrograph and EL image for CVD-grown polycrystalline and single-crystalline WSe_2 monolayers.

Owing to direct EL observations, we can perform spatial polarization-resolved EL spectroscopy to investigate detailed light-emitting properties. Figure 3 exhibits polarization-resolved EL spectra obtained at two different positions of single-crystalline WS_2 monolayer flake, in which each spectrum in Fig. 3 was recorded inside crystal and crystal edge regions, respectively. Only small EL polarization was obtained at lower temperature (< 40 K) inside crystal region (Fig. 3 bottom). It should be noted that the mixed contributions of two peaks is originating from both exciton and trion.

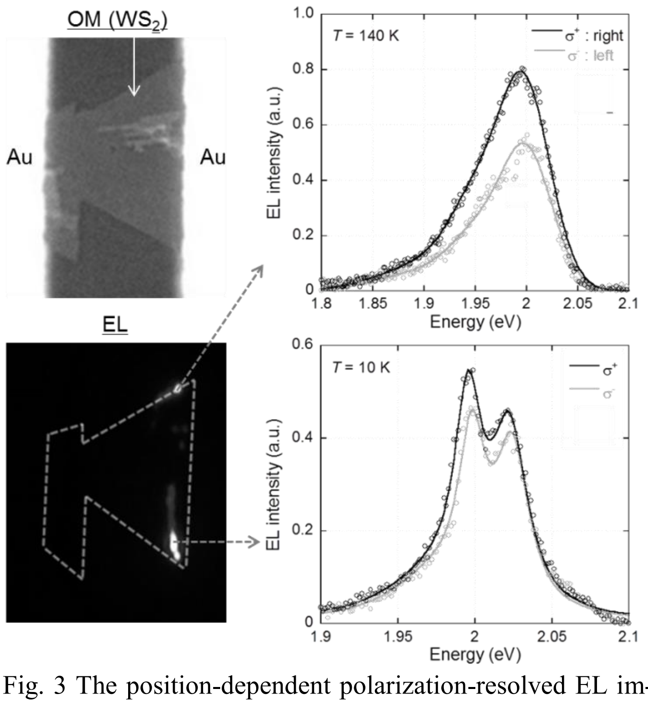

Fig. 3 The position-dependent polarization-resolved EL imaging and spectroscopy for single-crystalline WS_2.

In contrast, larger EL polarization was observed at higher temperature of > 100 K at crystal edge region (Fig. 3 top). 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 (PL) mapping done in same crystal. As a result, we founded out that the local strain induced at crystal edge regions due to lattice mismatch with substrate 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 next-generation optoelectronic applications.

3. Conclusions

We newly proposed a versatile and simple approach to fabricating TMDC light-emitting devices using electrolyte, and successfully observed their EL in various TMDCs. The simplicity of our light-emitting structure allows us to perform spatial EL polarization measurements. Consequently, we firstly observed robust circularly polarized EL near room temperature, arising from topological features of TMDCs. The established methods and evaluations open a route for designing practical chiral light sources based on monolayer TMDCs toward future quantum information technology.

Acknowledgements

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