Superacid Treatment on Transition Metal Dichalcogenides

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Abstract

Superacids are strong acids with an acidity higher than pure sulfuric acid. Recently, superacid treatment of monolayer transition metal dichalcogenide (TMDC) flakes, such as MoS2 and WS2, has shown a dramatic enhancement of optical properties, such as photoluminescence (PL) intensity. The superacid molecule is bis(trifluoromethane)sulfonimide (TFSI). In this review paper, we summarize and discuss the recent works and the current understanding of the TFSI treatment, and finally, we describe the outlook of the treatment on monolayer TMDCs.

1. Introduction

Since the isolation of graphene from graphite by a mechanical exfoliation method using Scotch tape was discovered, atomically thin layered materials with a few atoms thick have been studied extensively [1, 2]. Graphene has a linear band dispersion and is known as a semi-metallic material. Transition metal dichalcogenide (TMDC), which is the subject of this paper, is also an atomic-layered material comprising a thickness of three atoms, a sub-nanometer thick, and a direct band gap in the monolayer limit [3–6]. Due to their 2D confined semiconductive properties, TMDCs are attracting attention for electronic devices, such as atomic layer transistors, and as semiconductor materials for the construction of atomically thin optical devices via their direct band gap nature [6–10].

Representative TMDCs have a composition of MX2 (M = transition metals, X = chalcogen atoms) and are a group of crystalline materials whose structure has a transition metal ion sandwiched between two chalcogen atoms [3]. At the monolayer limit, the material is an ultrathin crystal with a layer thickness of about 0.7 nm [3]. It is possible to isolate monolayers from bulk MX2 crystals by either the mechanical exfoliation method or by growing via chemical vapor deposition (CVD) techniques [3]. Notably, the electronic state and optical properties change with the number of layers. In the multiple-layered samples, the edge of the valence band is at the Γ point, and the bottom of the conduction band is between the Γ and K point directions, resulting in an indirect band gap semiconductor [5, 6]. By contrast, at the monolayer limit, the valence band and the edge of the conduction band are located at the K point, making it a direct band gap [4–6]. The monolayers are expected to apply to light-emitting diodes (LEDs) [11, 12], solar cells [13], and photodetectors [14–17] from the direct band gap nature. In addition, monolayer TMDCs are transparent due to their atomically thin and flexible structures; therefore, TMDCs would also be a platform for innovative optoelectronic devices for transparent displays with a foldable design.

The bottleneck for applying optical devices is the low quantum yield (QY) of monolayer TMDCs. For example, the QY of monolayer MoS2 is reported to be between 0.01% and ~1% [4, 18–20]. WS2 has a relatively high QY, but it is still around several % [18, 21]. This review paper discusses the molecular chemistry technique known as superacid treatment to enhance the photoluminescence (PL) QY of monolayer MoS2 and WS2, and reviews the surface chemistry, display applications, and challenges in this molecular technique (figure 1).

2. Optoelectronic properties of TFSI-treated TMDCs

2.1. First experiment on exfoliated MoS2

2D-layered materials have shown profound physics as the materials thin down to the monolayer limit [24–27]. Emerging new physics, along with thickness thinning, has stimulated many future applications in electronics.
and optoelectronics, with superior performance that could surpass traditional semiconductors. Researchers have devoted significant efforts to studying light–monolayer interactions, including their absorption, scattering, and emission properties \[28–30\]. In the early stages of 2D research, researchers discovered that the PL of TMDC materials becomes stronger as the layer thickness decreases to a monolayer \[5, 31, 32\]. This unique feature has been observed in most 2D TMDC materials, such as MoS\(_2\), W\(_\text{S}_2\), W\(_\text{Se}_2\), and Mo\(_\text{Se}_2\) \[17, 21, 33, 34\]. The stronger PL signal in the monolayers was attributed to the indirect-to-direct bandgap transition as the thickness decreased to a monolayer \[35–37\]. In thin-layered 2D materials, researchers also noticed that their optical signal intensities (PL/Raman) were not merely determined by the materials’ intrinsic properties; they were also strongly affected by the dielectric environment \[38–42\]. It has been shown that both the in-coupling (absorption/excitation) and out-coupling (emission/scattering) of light–monolayer interactions are primarily controlled by surrounding structures and dielectrics \[43–48\]. External factors could lead to an enhancement or suppression of PL/Raman intensity up to a few orders of magnitude.

To further understand photon physics, it is essential to exclude the abovementioned external factors to obtain intrinsic material properties. In terms of optical properties, PL QY is a key metric used to evaluate the potential of the material for optoelectronic applications. PL QY is defined as the ratio of radiative recombination to the total recombination rate, which dictates the upper optoelectronic performance limits for LEDs, lasers, and other optoelectronic applications. Although the PL signal was stronger in TMDC monolayers than in multilayers, the quantitative measurement shows that the PL QYs were quite low (e.g., PL QYs of pristine MoS\(_2\) monolayers are typically <1%) \[22\]. The low PL QY of monolayers was initially attributed to their high defect density. Therefore, various approaches have been proposed, with efforts focusing on fixing defects \[49\]. It has been shown that PL QYs can be enhanced by depositing capping layers, applying chemical treatments, etc \[22, 50–53\]. One of the most effective approaches to improving PL QYs is superacid treatments, which use the bis(trifluoromethane)sulfonimide (TFSI) molecule to treat monolayer semiconductors. After the treatment, the PL QYs of TFSI-treated MoS\(_2\) monolayers were reported to be close to unity, where the enhancement of PL QY can be more than 2 orders of magnitude (figure 2). The minority carrier lifetime also increased from ∼50 ps to ∼10 ns, significantly suppressing the non-radiative recombination pathways. The highest PL QY enhancement was observed in a relatively low generation rate regime. The PL QY of the treated MoS\(_2\) monolayers showed a bimolecular droop at a higher generation rate, corresponding to a bi-excitonic annihilation process. The results reveal the excitonic nature of monolayer semiconductors from a pump power-dependent PL QY measurement based on recombination pathway analysis. Bimolecular decays of monolayer TMDCs observed at high pump power regimes were attributed to the biexciton annihilation process. The existence of the biexciton and excitonic natures in TMDCs was further confirmed by following-up studies \[54, 55\].

In a later study from the first TFSI treatment on a monolayer MoS\(_2\), the PL spectra of the treated MoS\(_2\) on different substrates were revealed to be unchanged; the researchers transferred an exfoliated and TFSI-treated...
monolayer MoS2 on quartz, sapphire, and strontium titanate (STO). The PL spectra are identical on each substrate. On the other hand, the time-resolved PL measurement revealed that the exciton-exciton annihilation rate constant was highly dependent on the substrate permittivity, an order of magnitude difference between quartz to STO at a high excitation rate \(56\).

2.2. Other exfoliated TMDCs and TFSI treatments

The surfaces of conventional semiconductors are crystal terminations, which are inevitably full of dangling bonds. When the semiconductor thins down to a few nanometers (e.g., 10 nm thin GaAs), it needs to be passivated with cladding layers (e.g., AlGaAs) to achieve a high PL QY \(57\), while the surfaces of ideal layered materials are self-terminated. Therefore, monolayer semiconductors are expected to exhibit a high PL QY. Unfortunately, the PL QY of TMDC monolayers was quite low, countering the expected high performance of the materials. After the first demonstration of TFSI treatment on MoS2, the TFSI treatments were also applied to other TMDC monolayers (figure 3) \(21, 53, 58–64\). The PL QYs of WS2 monolayers increased to near unity after the treatment, showing similar results as MoS2 monolayers. Notably, the treatment was ineffective on selenium-based TMDC monolayers, where the PL QY of the treated WS2 and MoSe2 monolayers showed a slight decrease in PL QYs by an order of magnitude or less. The changes in minority carrier lifetimes were consistent with the changes in the PL QYs. Sulfur-based TMDCs (MoS2 and WS2) showed increases in PL QY and lifetimes after TFSI treatment, while selenium-based TMDCs showed decreases in PL QYs and lifetimes after TFSI treatment. This observation was initially explained by the native defects present in different kinds of TMDC monolayers. Later studies have indicated that the mechanism of the TFSI treatment is effectively a doping treatment for monolayers \(65–67\). Various ionic chemicals acting as p-dopants were used to show pronounced PL enhancement in monolayer TMDCs (figure 3(b)) \(59\). The amount of enhancement is shown in figure 3(c). Given that the formation of excitonic species in TMDC monolayers is determined by background carrier concentrations, the doping effect of the TFSI treatment led to a modulation of excitonic populations \(67–71\). Therefore, changes in PL QYs were determined by the radiative recombination efficiency of the dominant excitonic

Figure 2. (a) PL images of a MoS2 monolayer before and after the TFSI treatment; (b) PL QY as a function of the generation rate and incident power for an as-exfoliated MoS2 monolayer. Reproduced with permission from \(22\). Copyright 2015 American Association for the Advancement of Science.

Figure 3. (a) Pump power dependence of PL QYs for as-exfoliated and chemically treated TMDC monolayers. Copyright 2016 American Chemical Society. (b) Structures of TFSI chemicals used for the treatments. (c) PL enhancements by different TFSI treatments on WS2 monolayers. Reproduced with permission from \(21, 59\). 2016 American Chemical Society, 2021 Nature Publishing Group.
species at the carrier concentration caused by TFSI doping. Table 1 summarizes the works of TFSI treatment. More details about the TFSI mechanism will be discussed in the following sections.

| Table 1. Studies of TFSI treatments on 1L-TMDs. |
|-----------------------------------------------|
| **TMDC materials** | **Type of acid treatment** | **Changes in optical properties** | **References** |
| 1L-MoS2 | TFSI | MoS2: PL QY 0.1% to 95% | [22] |
| 1L-MoS2 | TFSI through PMMA | MoS2: PL QY 0.1% to 95% | [50] |
| 1L-WS2 | TFSI | MoS2: PL QY 1% to 95% | [20] |
| 1L-MoS2 | TFSI | MoS2: PL QY 0.1% to 30% | [21] |
| 1L-WS2 | TFSI Salts | MoS2: PL QY 0.2% to 14% | [59] |
| 1L-MoS2 | TFSI | MoS2: PL QY 2.7% to 56.7% | [60] |
| 1L-WS2 | TFSI | MoS2: PL QY 0.1% to 9% | [38] |
| 1L-WSe2 | TFSI | MoS2: PL QY 2.7% to 56.7% | [60] |
| 1L-MoS2 | TFSI | MoS2: PL QY 1% to 95% | [20] |
| 1L-WS2 | TFSI | MoS2: PL QY 10% to 95% | [20] |
| 1L-WS2 | TFSI | MoS2: PL QY 3% to 1% | [51] |
| 1L-MoS2 | TFSI | MoS2: PL QY 9% to 1% | [52] |
| 1L-WS2 | TFSI | MoS2: PL QY 3% to 1% | [51] |
| 1L-MoS2 | TFSI | MoS2: PL QY 1% to 95% | [20] |
| 1L-MoS2 | UV-TFSI | MoS2: PL QY x 25 | [73] |
| 1L-WS2 | UV-TFSI | MoS2: PL x 250 | [74] |

2.3. CVD-grown samples

TFSI treatment was also applied to CVD-grown samples [20, 72, 73], which potentially enabled a large area of high luminescent materials. In the CVD-grown samples, the PL QY was not particularly high in the as-grown flakes (e.g., about 0.1% in the as-grown monolayer MoS2) [20]. As grown MoS2 had a tensile strain of about 1% in a previous report, the strained flake did not show PL enhancement after TFSI treatment [20, 75]. The QY rather decreased with the treatment procedure for the strained flake. After the strain was released by transferring the flake to another substrate, the PL QY dramatically changed after the TFSI treatment. In the case of the CVD-grown monolayer MoS2, after releasing the tensile strain, the PL QY increased by about 30% via TFSI treatment [20].

The CVD-grown MoS2 had a wide variety of defective sites [76, 77], and the PL QY was highly dependent on growth conditions [20]. In the sulfur-rich condition, corresponding to the high sulfur precursor temperature when MoS2 was grown in the CVD chamber, the PL QY was 1% in the as-grown sample (figure 4). The sulfur-deficient sample, which was made by growing at a low sulfur precursor temperature, showed a PL QY of about 0.1% under the same excitation condition. The TFSI treatment changed the PL QY for the sulfur-deficient sample by up to about 30%, but the sulfur-rich sample did not show QY enhancement (figure 4(b)) [20]. This result suggests a relationship with defective sites for a key chemical factor of the superacid TFSI treatment of monolayer MoS2.

PL imaging showed the uniformity of the TFSI treatment of the CVD-grown monolayer MoS2. Strong uniform PL was observed in the triangular flake after the TFSI treatment (figures 5(a)–(c)) [20]. The treatment procedure was to anneal the material on a hot plate for 5 min after TFSI treatment. Recently, additional UV irradiation after TFSI treatment was proposed for effective PL enhancement in monolayer MoS2 (figures 5(d)–(f)) [23, 74]. The UV irradiation process showed PL enhancement in both exfoliated and CVD-grown monolayer MoS2 [74]. In the CVD-grown flake, a bright PL was achieved, and the distribution of the PL intensity in the identical flake was approximately double. The enhancement factor was about two orders of magnitude from the as-transferred sample via the TFSI and UV irradiation combination processes. As shown in the reported results, TFSI treatment can dramatically enhance the PL intensity in monolayer MoS2, but the protocol to obtain the enhancement is still under debate, and a new approach for consistent enhancement is important in both exfoliated and CVD-grown samples.

3. Plausible mechanism

3.1. Protonation environment

Superacid is a strong protonating agent. The protonation environment is an important factor for strong PL enhancement in monolayer MoS2 and WS2. Previously, a simple acid treatment procedure was reported on
Exfoliated monolayer MoS2. Sulfuric acid (H2SO4) treatment changes the PL intensity of monolayer MoS2 by a factor of two via the treatment, and further annealing induces PL enhancement (figure 6) [61]. This annealing procedure increases the acid concentration by evaporating water from the surface. The PL signal was deconvoluted to signals from trions and excitons for the spectra of the as-exfoliated and the annealed for 40 min. The integrated PL intensity in figure 6(c) shows that both the trion and exciton signals increased with increasing annealing time. In this process, oxidation of the monolayer MoS2 was suggested [61]. The decay process of excitons, including the formation of trions and Auger recombination process, which would be suppressed by the oxidation process, would result in the PL intensity increasing via oxidation [61, 78, 79]. While, the PL intensity of trions would be maintained via oxidation processes, as suggested in the previous reports (both chemical oxidation [78] and electrostatic oxidation [79] processes). The reduction of the formation rate of the trion and Auger recombination process cancel each other out in the oxidation process. Therefore, in the protonation

Figure 4. PL QY for the CVD-grown monolayer MoS2 with various growth conditions. (a) The setup for the growth; (b) the PL QY for the as-transferred (released strain sample) and TFSI-treated flakes. Reproduced with permission from [20]. Copyright 2016 American Chemical Society.

Figure 5. (a)–(c) The uniformity of the PL intensity for the CVD-grown monolayer MoS2 in the original annealing protocol after the TFSI solution process; (d)–(f) UV-assisted process to enhance the PL intensity after TFSI treatment. Reproduced with permission from [20, 74]. Copyright 2016 and 2020 American Chemical Society.
environment shown in figure 6, an extra event would be incorporated in the trion PL enhancement process. A similar enhancement could not be observed in the case of Li2SO4 instead of the treatment with H2SO4 [61].

Similar behavior was also observed in the case of superacid TFSI treatment. Therefore, protonation is an important factor in enhancing the PL intensity of trions. The protonation and passivation of defective sites might happen in the acid treatments on monolayer MoS2.

There are several scenarios of surface chemistry have been proposed. The three protons bonded to a S-vacancy in a charged state with −1 may be a scenario with no deep-level state in the band gap. The natural n-doped state in MoS2 may generate the situation of the −1 state under the strong protonation environment of TFSI treatment [63]. In another scenario, O2 interaction on the S-vacancy is also proposed [63]. This situation may happen because TFSI treatment procedures are done in ambient environments. Treatment in the ambient condition is critical to show the strong PL intensity in the TFSI treatment process [74]. TFSI treatment may clean

Figure 6. Simple acid (H2SO4) treatment on monolayer MoS2 and PL enhancement in the annealing process. XA and XA are PL signals of exciton and trion. Reproduced with permission from [61]. Copyright 2018 American Chemical Society.
the S-vacancies, and consecutive $O_2$ interaction may happen. In another case, additional UV irradiation makes the strong PL intensity; in the process, the OH passivation at S-vacancies is a candidate. In this case, although the density of states remains near the conduction band edge, the mid-gap states are removed. Various research with superacid molecules has been reported, but the surface chemistry has not been concluded yet.\[80\]. Because the chemistry on the surface is complicated, it requires more research to conclude.

3.2. Surface encapsulation

The strong PL intensity in monolayer MoS$_2$ and WS$_2$ was observed from the dried surface after the TFSI solution process. The drying procedure included blowing the TFSI solution using dry nitrogen gas. As shown in figure 7(a), the dried surface showed a strong PL intensity; however, the brightness decreased dramatically under a TFSI aqueous solution (figure 7(b)) [23]. The concentration in the in situ measurement of the TFSI solution was the same as the initial treatment. Further drying of the surface regenerated the strong PL intensity in the air (figure 7(c)). Drying the surface after TFSI treatment is the key process for enhancing PL intensity. In the case of the aqueous solution, TFSI molecules interact well with the bulk water because of their hygroscopic character and because they have difficulty interacting with the surface of the monolayers (figure 7(d)). As a result, the PL intensity remained small. The atomic force microscope (AFM) image of the dried surface shows a nm-thick film forming on MoS$_2$ selectively (not observed on the SiO$_2$ surface, as shown in figure 7(e)). The TFSI molecule is known to have a hygroscopic character, and the adsorption of water molecules from the environment would make the nm-thick film and form a highly concentrated superacid TFSI molecular environment on the monolayer MoS$_2$ surface. Importantly, a large portion of water is not appropriate for the TFSI treatment process; the PL intensity of the TFSI-treated monolayer MoS$_2$ decreases when increasing the amount of water in the treatment solution because the TFSI layer is not preferentially constructed. By keeping the TFSI thin layer enwrapped with polymethyl methacrylate (PMMA) polymer, the strong PL intensity can be maintained for more than 30 days in the air (figures 7(f) and (g)) [23]. The encapsulation of the monolayer surface with a thin layer of TFSI molecules would be the key physicochemical factor in enhancing PL intensity. The results also indicate the importance of the protonation environment on the surface of monolayers to enhance PL intensity.

Figure 7. Surface encapsulation with a thin layer of the TFSI molecules on the dried monolayer MoS$_2$ in the air after the TFSI solution treatment. The PL mapping images of (a) just after treated and measured in the air, (b) immersion in TFSI aqueous solution, (c) re-dried surface. (d) TFSI molecules interact with MoS$_2$ surface when the small water content is in the solution. (e) AFM image of the TFSI-treated MoS$_2$ surface. (f, g) Enwrapping with PMMA polymer is effective in keeping the TFSI treatment for 30 days in the air. Reproduced with permission from [23]. Copyright 2021 American Chemical Society.
3.3. Doping effect

TFSI molecule is acting as a p-type agent. The doping effect on TMDCs (equivalent to p-type doping) derives from the mechanism of surface charge transfer \[59, 65, 72, 81, 82\]. To understand the doping effects on TMDC monolayers, the researchers performed a control experiment using an untreated MoS\(_2\) monolayer over a metal-oxide-semiconductor (MOS) capacitor structure to mimic the doping effect of TFSI treatment (figure 8(a)) \[65\]. Such device structure is widely used to examine the exciton species present in different types of 2D materials as it can vary the background carrier concentration simply by applying a gate voltage \[69 – 71, 79, 83, 84\].

In this experiment, the monolayer semiconductors were electrostatically doped by applying a voltage across the gate oxide. Meanwhile, the change in PL QY as a function of the electrostatic doping level was examined. The excitons formed from photogenerated carriers evolved into different types of excitonic species. The formed excitons were neutral when the doping level was low (close to intrinsic). When the doping level was high, the excitons became positive or negative trions, depending on whether the doping was p-type or n-type, respectively \[68, 69, 71, 79, 85 – 87\]. The trions had a lower radiative recombination rate than the neutral excitons \[79, 88\]. As a result, the PL QY reached close to unity when MoS\(_2\) and WS\(_2\) monolayers were electrostatically doped by applying a voltage across the gate oxide. Meanwhile, the change in PL QY as a function of the electrostatic doping level was examined. The excitons formed from photogenerated carriers evolved into different types of excitonic species. The formed excitons were neutral when the doping level was low (close to intrinsic). When the doping level was high, the excitons became positive or negative trions, depending on whether the doping was p-type or n-type, respectively (figure 8(c)) \[68, 69, 71, 79, 85 – 87\]. The trions had a lower radiative recombination rate than the neutral excitons \[79, 88\]. As a result, the PL QY reached close to unity when MoS\(_2\) and WS\(_2\) monolayers were electrostatically doped from an initially electron-rich state to an intrinsic state (figures 8(d) and (e)) \[89 – 91\]. Figure 8(d) shows that the population changes from the trion to exciton as sweeping the gate voltage from positive to negative. It also shows that the recombination process in trion is predominately nonradiative while in exciton it is mostly radiative. An effect of the electron counter-doping nature of the TFSI molecule made the excitons neutral, leading to near-unity PL QY. This mechanism was further confirmed in the TFSI-treated monolayer transistor, where a threshold voltage shift was observed \[22, 72, 81\]. The decrease in PL QY after TFSI treatment in WSe\(_2\) and MoSe\(_2\) monolayers was also in agreement with the observation seen in the gated PL measurements, where positive trions were formed in those initially intrinsic materials with relatively smaller bandgaps.

The doping effect of TFSI treatment as a function of the encapsulation layer’s thickness is discussed in a previous study \[50\].
This paper reviewed the superacid TFSI treatment procedures on monolayer MoS$_2$ and WS$_2$ to enhance PL intensity and corresponding QY. The plausible physicochemical mechanism is also discussed by forming an nm-thick TFSI layer, which would effectively exhibit the protonation environment and doping effect on the monolayer flakes. The prospective applications in these bright monolayers would be transparent optical devices, such as display applications. We described the AC-voltage modulated display device from the TFSI-treated monolayer WS$_2$ devices show EL efficiency of $\sim 1.2\%$ (figure 9(b)). Notably, the efficiency is limited by the PL QY efficiency droop in a high-injection regime. The so-called ‘biexciton annihilation’ process causes an efficiency droop, which has been widely observed in TMDC monolayers (figure 9(c)) [18, 54, 93, 94]. Recent reports have shown that by introducing mechanical strain, the electronic band structures are significantly changed, which could potentially suppress biexciton annihilation [66]. The strain concept demonstrates a promising route to achieving a highly efficient and high-power emitter for light-emitting and laser applications.

4. Device applications

The discovery of TFSI treatment has attracted much attention because the resulting nearly perfect performance of the treated monolayers could potentially be used to develop highly efficient LEDs, lasers, and solar cells. However, device performance regarding TFSI treatment has rarely been reported due to constraints related to the treatment, such as process incompatibility. Several studies have shown that the effect of TFSI treatment can be removed during device fabrication processes. The encapsulation strategies, as discussed in the previous sections, were developed to prevent the removal of the effect [50]. Another constraint, along with the enhancement of PL QY, is the increase in the electrical resistance of TMDCs due to the counter-doping effect. As a result, the semiconductor becomes intrinsic, and the injection current is limited, hindering their optoelectronic performance. In terms of electrical devices, the counter-doping effect leads to a threshold voltage shift in transistor characteristics (figure 9(a)) [22, 72]. Note that some reports have shown that the subthreshold swing of the treated devices also improved, which was attributed to a reduction in trap density due to TFSI treatment. In terms of LEDs and lasers, the trade-off between PL QY and injection current largely limits the brightness and efficiency of a TMDC-based device. A new device scheme was proposed to overcome the carrier injection issue using a MOS capacitor structure (figure 9(d)) to drive electroluminescence (EL) [10, 11, 92]. It is a capacitive structure where TMDCs are laterally contacted to the source electrode atop the gate oxide. Meanwhile, AC voltage was applied between the gate and the source contacts. TFSI-treated WS$_2$ devices show EL quantum efficiency of $\sim 1.2\%$ (figure 9(b)). Notably, the efficiency is limited by the PL QY efficiency droop in a high-injection regime. The so-called ‘biexciton annihilation’ process causes an efficiency droop, which has been widely observed in TMDC monolayers (figure 9(c)) [18, 54, 93, 94]. Recent reports have shown that by introducing mechanical strain, the electronic band structures are significantly changed, which could potentially suppress biexciton annihilation [66]. The strain concept demonstrates a promising route to achieving a highly efficient and high-power emitter for light-emitting and laser applications.

5. Summary and outlook

This paper reviewed the superacid TFSI treatment procedures on monolayer MoS$_2$ and WS$_2$ to enhance PL intensity and corresponding QY. The plausible physicochemical mechanism is also discussed by forming an nm-thick TFSI layer, which would effectively exhibit the protonation environment and doping effect on the monolayer flakes. The prospective applications in these bright monolayers would be transparent optical devices, such as display applications. We described the AC-voltage modulated display device from the TFSI-treated bright monolayer flakes. Currently, widespread research has focused on TFSI treatment, but the surface chemistry of the mechanism requires more in-depth research. One probable scenario would be defect passivation of sulfur vacancies by sulfur adatoms [60, 63, 95] reported by analysis via transmission electron microscope [60, 95]; conversely, PL behavior indicates the generation of defective sites even after the TFSI treatment process, which is identified by the bound-exciton-related signal at low temperatures [23]. Or TFSI treatment procedure may be concluded as a simple doping effect between the TFSI molecules and TMDCs. Additionally, the TFSI process is effective only for sulfide-based materials, such as MoS$_2$ and WS$_2$, not for MoSe$_2$ and WSe$_2$ [21]. It is still difficult to explain why selenium-based materials do not show bright PL after TFSI treatment. Although the TFSI-treated samples showed strong systematic enhancement in PL intensity, and the display device was demonstrated [10], critical applications in science (physics and chemistry) and engineering
are still missing. Encapsulation of thick polymer layers, such as PMMA, can maintain the bright PL of the TFSI-treated monolayers [23], but more stability in air, especially in a vacuum, would be essential for exploring new optoelectronic science. Under vacuum conditions, the PL intensity decreases, even after the encapsulation caused by the meshed porous structure of the wrapped PMMA layer. The water (and/or TFSI) molecules in the thin TFSI layer at the interface of monolayer TMDGs would evaporate through the porous structure. Thus, the TFSI acidic film would no longer be effective. The strong PL behavior might be effective for generating highly efficient photovoltaics and optical sensors at nm-thick active layers; however, the strong acidity of the TFSI treatment process may damage the metal electrodes of the devices, and the TFSI molecular layers on monolayers (shown in section 3.2) might interrupt the fabrication process. More trials in various fields would be effective for further investigations of the bright atomically thin material, and the significant game-changing phenomenon and devices would be on par with in-depth research.

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Data availability statement

No new data were created or analysed in this study.

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