Aggregation-Induced Emission Luminogens for Direct Exfoliation of 2D Layered Materials in Ethanol

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Aggregation-induced emission (AIE) luminogens are an important type of advanced functional materials with fantastic optical properties and have found potential applications in organic electronics, biochemistry, and molecular imaging. Herein, this article presents a novel application of AIE luminogens (AIEgens) for efficient exfoliation of layered transition metal dichalcogenides (TMDs, such as MoS2 and WSe2). From the 1H NMR spectroscopic analysis, the designed AIEgens can insert into the space between layers of MoS2 in ethanol solution and the dynamic molecular rotation against the weak interactions affords large-scale few-layer MoS2 nanosheets (7–8 layers) with enhanced smoothness. The 3D AIEgens play a significant role in preserving the crystal lattice of MoS2 even at high pressure (>15 GPa). More importantly, the new approach can also be used for exfoliation of WSe2 to achieve large-scale few-layer nanosheets. The present work thus provides a facile and high-yielding synthetic method for accessing on a large scale 2D layered materials with enhanced properties for high-technology applications.

1. Introduction

2D layered materials with a mono or limited number of layers are receiving growing interest because of their unique electronic and optical properties.[1–3] Molybdenum disulfide (MoS2) is a member of the layered materials family, and possesses a narrow indirect bandgap (~1.2 eV) in bulk, and for which the nature of the bandgap from indirect to direct can be tuned by reducing the thickness.[4,5] These thickness-dependent properties of the layered MoS2 can dictate the potential applications for electronic devices, photocatalysis, chemsensors, spintronics, etc.[6,7] For example, the photoluminescence spectra of monolayered MoS2 reveal a dramatic increase in luminescence efficiency compared to multilayered and bulk MoS2 systems.[8]

Up to now, much effort has been devoted to developing facile, reproducible, and highly efficient methods to achieve high-performance mono-/few-layer MoS2. Historically, with respect to graphene, monolayer MoS2 can also be accessed from bulk powders by mechanical exfoliation albeit in low yield.[9] Chemical vapor deposition (CVD) is a general approach for the preparation of 2D materials, but in practice, CVD-synthesized monolayers remain scant.[10] Zhang and coworkers reported a controllable electrochemical lithiation method for the fabrication of high-yield, monolayer MoS2, but this electrochemical lithiation process is considered to be an uncontrollable method for exfoliation of MoS2, given that by-products, namely Mo nanoparticles and Li2S appear on the surface of the MoS2.[11] Liquid exfoliation of bulky layered MoS2 is considered as one of the most effective techniques to produce high-yield and high-quality monolayer flakes.[12] However, the intrinsic properties of solvents, such as polarity, intermolecular interactions, and solubility parameters limit the productivity using liquid exfoliation for 2D layered materials.[13] Kalantar-zadeh and coworkers investigated ten organic solvents for the grinding-assisted ultrasound exfoliation of MoS2, and concluded that the category of solvent plays a significant role in the improvement of the concentration of exfoliated suspensions.[14] Vaia and coworkers systematically studied the mechanism of the solvent-assisted exfoliation of MoS2 in N-methyl-2-pyrrolidone (NMP) and found both the O2 content and the fraction of water in the NMP were controlling factors when optimizing the yield of MoS2 flakes.[15] Zhang and coworkers have successfully exfoliated layered MoS2 with the highest concentration of MoS2 nanomaterials at 0.018 ± 0.003 mg mL−1 in a mixture of ethanol/water (f(w) = 45%). By contrast, in pure ethanol or water, only a low concentration of MoS2 was achieved.[16]
Besides solvent-assisted exfoliation techniques, additive-assisted techniques have been widely used for the exfoliation and dispersion of 2D materials in solution. This is typically employed for bulk transition metal dichalcogenides (TMDs) and graphite, and the important thing is that the additive@MoS$_2$ can be directly integrated for application without further treatment.$^{[26]}$ Huang and coworkers reported a facile and scalable method to achieve mono- and few-layer nanosheets of bulk transition metal dichalcogenides by employing poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), which can be utilized for high-performance organic solar cells as a hole extraction layer.$^{[18]}$ Furthermore, both the biomolecule-assisted and surfactant-assisted exfoliation of layered MoS$_2$ can play important roles in achieving stable dispersions of atomically 2D layered materials in water.$^{[19,20]}$ It has become clear that the solvent exhibits little effect on the exfoliation of layered MoS$_2$ in additive-assisted systems.

Tang discovered an abnormal optical phenomenon, known as aggregation-induced emission (AIE) in 2001.$^{[21]}$ Representative luminogens, such as tetraphenylethylene (TPE) and hexaphenylsilole (HPS), exhibited negligible emission in solution but enhanced emission in the solid state. Furthermore, the working mechanism for AIE can be visualized in terms of the restriction of intramolecular motion (RIM), in which the terminal phenyl ring groups were dynamically rotated against the central core as a rotor in solution, and the photonic energy transferred to thermal energy via a nonradiative process. In contrast, molecular rotation was restricted and thus it emitted fluorescence in the aggregation state.$^{[22–25]}$ This novel molecular rotation model based on AIEgens inspired us to develop potential applications in the 2D semiconductor field.

The 2D single-layer MoS$_2$ is bound by strong in-plane bonding (S-Mo-S) with a thickness of 0.6–0.9 nm.$^{[26]}$ and the bulk MoS$_2$ is assembled by numbers of 2D single-layer MoS$_2$ via weak van der Waals forces with a considerable space between layers.$^{[27]}$ Zhang et al. reported that alkali metal,$^{[28]}$ alkali naphthalenide compounds$^{[29]}$ can insert into the interlayer spaces of the bulk TMDs for achieving mono- or fewer layer 2D TMDs. According to the RIM mechanism, is it possible that AIEgens also can insert into the interlayer spaces and break the weak intermolecular interactions and exfoliate the 2D layered material in solution through the molecular rotation process? With this in mind, a feasible and efficient AIEgens-assisted strategy for exfoliation of layers 2D layered materials is described herein. Indeed, our work indicates that the AIEgens play a crucial role in exfoliating layered MoS$_2$ and WSe$_2$. Meanwhile, it can improve the yield on a large scale and afford high quality few layer MoS$_2$/WSe$_2$ flakes. More importantly, the exfoliated MoS$_2$ containing the AIE was composed of a crystal lattice that was stable even at pressures as high as 15 GPa. We believe this methodology provides a strategy to develop a new generation of semiconductor devices for potential applications under hyperbaric environments.

2. Results and Discussion

2.1. Synthesis

According to the synthetic route shown in Figure 1, the compound TPENA (Figure 1a) was synthesized by the Suzuki–Miyaura coupling of 1,8-dibromonaphthalene and 4-(1,2,2-triphenylvinyl) phenylboronic acid under Pd-catalyzed conditions in 70% yield. The molecular structure of TPENA was fully characterized by $^1$H/$^1$C NMR spectroscopy and high resolution mass spectrometry. The TPENA exhibits good solubility in common organic solvents (such as dichloromethane, tetrahydrofuran (THF), and toluene), and considerable solubility in ethanol, but is insoluble in water. The optimized geometries of the TPENA molecule was performed by density functional theory (DFT) using Gaussian 09 program package.$^{[30]}$ The two TPE moieties were introduced at the 1,8-positions of the naphthalene ring giving a crowded space configuration, and both TPE moieties are near perpendicular to each other; one of the TPE groups is parallel to the naphthalene ring. The dimensions of the TPENA are ≈6.71 Å, ≈15.20 Å, and 4.96 to 11.58 Å along the x-, y-, and z-axes, respectively (Figure S5, Supporting Information).

2.2. AIE Properties

The photoluminescence (PL) spectrum of TPENA was investigated in a mixture of THF/water using different water fractions ($f_w$), and was found to emit a weak blue emission peak at 458 nm in solution with low quantum yield ($\Phi_f < 0.05\%$), as

Figure 1. a) Synthetic route to molecule TPENA and b) the optimum structure of TPENA.
shown in Figure 2a. On increasing the water fraction to 60% (Figure 2b), the PL intensity only changed slightly. Further, the emission was enhanced by continuously increasing the fraction of water, and the maximum emission intensity observed was at \( f_w < 99\% \) with an 80-fold enhancement and much higher quantum yield (\( \Phi_f \approx 0.67 \)) versus that in pure THF solution. Thus the TPENA is clearly exhibiting features associated with AIE (Figure S6, Supporting Information).

2.3. Exfoliation of 2D Layered Materials

According to previous reports, ethanol has been widely exploited to exfoliate 2D layered materials. Herein, 1 mL ethanol was added to a mixture of MoS\(_2\) powder (0.5 g) and TPENA (2.5 mg), which was further grinded in a mortar and pestle for 30 min. After grinding, the sample was sonicated in ethanol (20 mL) for 1 h followed by centrifuging twice at 5000 rpm min\(^{-1}\). The suspension containing MoS\(_2\) nanosheets was then collected and characterized by UV–vis, scanning electron microscope (SEM), transmission electron microscopy (TEM), atomic force microscopy (AFM), and Raman spectroscopy. For further comparison of the effect of the AIE molecule, the thickness of the exfoliation sample was found to be more than 40 layers Figure S10B (Supporting Information); the thickness of the monolayer MoS\(_2\) was a constant value of \( \approx 0.6–0.9 \) nm\([26,31]\). The exact values of the thickness was fitted according to the multiple scans and histogram using AFM on MoS\(_2\), and the height distribution histogram shows that the corresponding thickness of the AIE-assisted exfoliated, grinding-assisted and sonication-assisted exfoliated MoS\(_2\) nanosheets were uniformly dispersed with average layers ranging from 7–8 layers, 38–40 layers, and 50–53 layers, respectively.

Thus, both the TEM and the AFM images indicate that the AIEgen is favorable for exfoliating the bulk MoS\(_2\) into high quality ultrathin nanosheets. While in the absence of the AIE molecule, the thickness of the exfoliation sample was found to be more than 40 layers Figure S10B (Supporting Information); On the other hand, without grinding, the AIEgens-assisted MoS\(_2\) sample displays a larger-scale, thicker nanosheets compared to TPENA@MoS\(_2\) samples (Figure S12, Supporting Information). More importantly, the AIE-active molecules of TPENA are adhered to the planar MoS\(_2\) flasks (shown as a light spot in Figure 3c1 and Figure S7 (Supporting Information)). Thus, we

![Figure 2. A) Emission spectra of TPENA in THF/water mixtures with different water fractions (\( f_w \)). B) Plots of \( I/I_0 \) values versus the compositions of THF/water mixtures of TPENA, in which \( I_0 \) is the PL intensity in pure THF solution. Inset: digital photographs of TPENA in THF and in THF/water mixtures (\( f_w = 90\% \)) taken under UV irradiation (\( \lambda_{ex} = 365 \) nm).](https://www.advancedsciencenews.com/doi/abs/10.1002/admi.202000012)
Note here that the AIEgen can be used for the effective exfoliation of layered MoS2 in ethanol, to achieve single- or few-layered MoS2 nanosheets.

The UV–vis absorption spectra of TPENA and the exfoliated suspension of MoS2 flakes via the three methods (TPENA@MoS2, grinding@MoS2, and sonication@MoS2) were also recorded. For TPENA, a maximum absorption band was observed at 337 nm in dilute THF (10 × 10⁻⁶ m), which is red-shifted by ~28 nm compared to naked TPE (λabs = 309 nm) (see Figure S18 in the Supporting Information);[32] this can be attributed to the expanding π-conjugation of the naphthyl group.[33] Whereas the dispersion of MoS2 nanosheets only exhibited a large red-shift of the broad and excitonic peak at 723 nm, which is attributed to the K point of the Brillouin zone.[34] The red-shift of the absorption peak edge is related to the size of the nanosheets.[35,36] In addition, the TPENA exhibited no obvious absorption behaviour beyond 337 nm, indicating that the absorption band at 723 nm for TPENA will have a limited effect on MoS2. The concentration of dispersed material is confirmed using the Lambert-Beer law and the extinction coefficient at 600 nm ε600 = 2104 mL (mg mg⁻¹)[37] with the concentration of the three samples being 0.03, 0.024, and 0.015 mg mL⁻¹ for TPENA@MoS2, grinding@MoS2, and sonication@MoS2, respectively. Obviously, grinding can crush the bulk MoS2 and improve the yield of nanosheets, but more importantly, the AIEgen-assisted additive technique plays a significant role to exfoliate and produce highly dispersible MoS2 flakes in ethanol with high efficiency and via a readily scalable method. Moreover, in the presence of TPENA, the duration of sonication required was shortened by 8–48 times without any reduction in yield compared to the reported method.[38]

2.4. Possible Exfoliation Mechanism

To gain insight into the exfoliation process, the interaction between molecular AIE and MoS2 was further investigated by ¹H NMR spectroscopy. The ¹H NMR spectra of TPENA and TPENA@MoS2 are illustrated in Figure 4. The ¹H NMR spectra of TPENA in d-methanol exhibited three groups of overlapping peaks between 7.10–7.20 ppm (triplet peak at δ = 7.10 ppm, a doublet at δ = 7.14 ppm, and a doublet of doublets at δ = 7.20 ppm). Interestingly, the ¹H NMR spectra of dispersions of TPENA@MoS2 are very different in d-methanol in comparison to that of pure TPENA. Although there are no additional resonances in the range 7.10–7.20 ppm, there are two distinct downfield chemical shifts for the doublet proton peak and multiple proton peak which appear at δ = 7.34 ppm and δ = 7.47 ppm, respectively. Although it is difficult to identify the proton peaks of TPENA, there is no doubt that the downfield shifts of the proton signals (δ = 7.34 ppm and δ = 7.47 ppm) are the result of de-shielding of the phenyl rings of TPENA by the large...
MoS₂ nanosheets. Moreover, one upfield shifted proton peak δ = 701 ppm is observed, which corresponds to the shielding of the proton peak of the TPE fragment.

Thus, we infer that the TPENA is well dispersed in MoS₂ with small content (0.5 g MoS₂ powder and 2.5 mg TPENA) in the whole grinding process, and the TPENA has more opportunities to intercalate into the space of pristine MoS₂; on the other hand, due to the concentration of TPENA in ethanol solution (20 mL) is very low (1.58 × 10⁻⁴ mol L⁻¹), the TPENA is almost dissolved to be a single molecular with free motion, which is easy to insert into the sandwiched structure of layered-MoS₂. Based on RIM mechanism, the phenyl group can freely rotate in ethanol solution, combining to the optimized molecular structure (DFT/B3LYP/6-31G*) and the ¹H NMR spectroscopic experimental observations, it is reasonable to propose that the naphthyl ring and one of TPE moiety of TPENA have inserted into the gap between layered MoS₂ with a spontaneous molecular motion, which would play a significant role in disrupting the weak intermolecular interactions and thereby exfoliating layered MoS₂. The possible mechanism for exfoliate layered MoS₂ as exploited herein is depicted as a schematic in Figure 4B.

Furthermore, to verify the feasibility of the above-mentioned method, we attempt to exfoliate the layered WSe₂ by following the same experimental procedure. Theoretically, the thickness of the monolayer WSe₂ is similar to MoS₂ at 0.6–0.9 nm.[19] From the AFM images of TPENA@WSe₂ in Figure 5, we found that the thickness of the WSe₂ nanosheets ranged from 5.86 to 6.17 nm with regular sizes in the micron range, which indicated that the TPENA@WSe₂ flakes also consisted of 7–8 layers with a regular smoothness in the dispersion (Figure S13, Supporting Information). The TPENA@WSe₂ nanosheets were further characterized by TEM images (Figure 5B) and confirmed by elemental mapping images (Figure 5D,E), scanning electron microscope (SEM) (Figure S14, Supporting Information), as well as Raman spectra (Figure S15, Supporting Information). While the controlled samples WSe₂ without the AIEgens-assisted technique consist of multilayer structures. (Figure S16, Supporting Information) In addition, without grinding, the thickness of AIEgens-assisted exfoliated WSe₂ nanosheets is in a range from 12.11 to 23.98 nm, meaning the grinding process would help intercalation. (Figure S17, Supporting Information)

2.5. Pressure-Dependent Lattice Vibrational Properties

Raman spectroscopy as a nondestructive diagnostic tool is widely applied to examine the phonon vibrations and crystalline structure of MoS₂. The mono-/few-layer MoS₂ exhibited two clear Raman peaks, which correspond to the interlayer vibrations (E₂g¹ mode) of the Mo and S atoms, and the interlayer vibrations (A₁g mode) of the S=S atoms against the Mo atom, respectively.[40] More importantly, the number of layers of MoS₂ can be determined using Raman spectra via measuring the distance between the A₁g and E₂g modes.[40] The Raman spectra of the TPENA@MoS₂ nanosheet was measured and bands at 380.8 and 405.6 cm⁻¹ were observed, indicative of few-layer MoS₂ which is consistent with the AFM results.

The electronic structure and lattice vibrational dynamics were investigated under high pressure by applying a hydrostatic pressure using a diamond anvil cell (DAC), with results shown in Figure 6. As the pressure increased from ambient to 25 GPa, both vibration modes were red-shifted to high wavelength from 380.8 to 3974 cm⁻¹ for the E₂g¹ mode, and 405.6 to 4374 cm⁻¹ for the A₁g mode with an increased intensity ratio of A₁g/E₂g¹ in addition, the intensity of the E₂g¹ mode almost disappeared (406.3 cm⁻¹) under high hydrostatic pressures (>25 GPa), due to the effect of normal compressive strain on the TPENA@MoS₂ nanosheet. However, the A₁g mode remained dominant (456.0 cm⁻¹). Without TPENA, the intensity of the E₂g¹ mode diminished somewhat when the hydrostatic pressures applied was up to 9 GPa (Figure S19, Supporting Information). A possible explanation is that the structural distortion would occur under high pressure, while the TPENA molecules between the two layers of exfoliated MoS₂ play a significant role to preserve the crystal lattice of MoS₂ even at the high pressure.

3. Conclusion

In this work, we present an efficient approach to exfoliate layered MoS₂/WSe₂ in the presence of AIEgen in pure ethanol. The obtained MoS₂ nanosheets exhibited a large-scale few-layer (7–8 layers). The AIE molecule TPENA plays a crucial role in improving the yield of exfoliating bulk MoS₂ over a shorter
sonication time. The \(^1\)H NMR spectra indicates that part of the TPENA molecule inserts into the gaps in layered MoS\(_2\), and the nature of the AIE molecule with its spontaneous molecular motion can disrupt the weak intermolecular interactions present in layered MoS\(_2\). In addition, the MoS\(_2\) nanosheets containing TPENA play a significant role to preserve the crystal lattice of MoS\(_2\) even under high pressure. Thus, this report presents a common approach for preparing 2D layered material (such as MoS\(_2\), WSe\(_2\)) nanosheets on a large scale and as thin-layered flakes and holds great potential for the utilization in strained semiconductor device applications, and some related work also is ongoing in our laboratory.

![Figure 5](image_url)

**Figure 5.** A) Atomic force microscopy (AFM) image of TPENA@WSe\(_2\) nanosheets, B,C) HRTEM images of TPENA@WSe\(_2\) nanosheets, and D–F) the corresponding element mapping images by energy-dispersive X-ray spectroscopy.

![Figure 6](image_url)

**Figure 6.** A) Pressure-dependent lattice vibrational properties of TPENA@MoS\(_2\) nanosheets and B) pressure-dependent Raman frequencies with variation in pressure.
4. Experimental Section

Materials and Methods: Unless otherwise stated, all reagents used were purchased from commercial sources and were used without further purification. $^1$H/$^{13}$C NMR spectra were recorded on an ADVANCE III HD 400 MHz NMR spectrometer and referenced $\delta = 7.26$ and 77.0 ppm for chloroform-D solvent with SiMe$_4$ as an internal reference, respectively; $\delta$-values are given in Hz. High-resolution mass spectra (HRMS) were taken on a TSQ Endura mass spectrometer operating in a MALDI-TOF mode. The surface morphologies of prepared samples were investigated using scanning electron microscopy (SEM, FEI Nova230) and transmission electron microscopy (TEM, FEI, Talos F200S), and the thickness of exfoliated nano-sheet MoS$_2$ was analyzed at atomic force microscopy (AFM, Bruker Dimension Edge SPM). Raman spectra were recorded at LabRAM HR 800 using a 532 nm visible-range laser.

Synthetic Procedures—Synthesis of 1,8-bis(4-(1,2,2-triphenylvinyl)phenyl)naphthalene (TPENA): A mixture of 1,8-dibromonaphthalene (0.51 mmol, 1.0 eq) and 4-(1,2,2-triphenylvinyl)phenylboronic acid (1.53 mmol, 3 eq) in toluene (15 mL) and ethanol (4 mL) at room temperature was stirred under argon, then potassium carbonate (2.04 mmol, 4.0 eq) and tetrakis(triphenylphosphine)palladium(0) (0.1 eq) were added. After the mixture was stirred for 30 min at room temperature under argon, the mixture was heated to 90 °C for 48 h with stirring. After cooling to room temperature, the mixture was quenched with water, extracted with CH$_2$Cl$_2$ (3 × 100 mL), washed with water and brine. The organic extracts were dried with MgSO$_4$ and evaporated. The residue was purified by column chromatography eluting with (CH$_2$Cl$_2$/hexane) to give target compound. $^1$H NMR (400 MHz, CDCl$_3$) $\delta = 7.91$ (dd, $J = 8.2$, 1.2 Hz, 2H), 7.58-7.49 (m, 2H), 7.40 (dd, $J = 7.2$, 1.3 Hz, 2H), 7.31-7.29 (m, 4H), 7.26 – 7.05 (m, 26H), 6.78 (d, $J = 8.5$ Hz, 4H), 6.73 (d, $J = 8.5$ Hz, 4H). $^{13}$C NMR (100 MHz, CDCl$_3$) $\delta = 144.45, 144.24, 144.18, 141.90, 140.91, 140.66, 140.27, 140.09, 135.40, 132.21, 131.67, 131.43, 131.36, 130.74, 128.95, 128.56, 127.98, 127.75, 127.61, 126.58, 126.36, 125.06 ppm; HRMS (MALDI–TOF): m/z called for C$_{62}$H$_{44}$ 789.0340, found 788.3428.

Synthetic Procedures—Exfoliation of Layered TMDs: According to reported procedure,[1][14] (TPENA@MoS$_2$) the general procedure for exfoliation of layered bulky TMDs solids followed the procedure below: 1 mL ethanol was added into 0.5 g MoS$_2$ powder (Sigma-Aldrich, $\approx 6 \mu$m) and 2.5 mg dye TPENA, the mixture was grinded in a mortar and pestle for 30 min. (Grinding@MoS$_2$) 1 mL ethanol was added into 0.5 g MoS$_2$ powder (Sigma-Aldrich, $\approx 6 \mu$m), the mixture was ground in a mortar and pestle for 30 min. (Sonication@MoS$_2$) 1 mL ethanol was added into 0.5 g MoS$_2$ powder without grinding and was kept at room temperature 30 min, then the solvent was evaporated. The three samples were then redispersed in 20 mL of ethanol and then sonicated for 1 h, while the temperature was maintained at 25°C by use of a water-cooled bath. To completely remove the bulk powder, firstly, the solution was centrifuged at 5000 rpm (Boeco, C-28A) for 1 h and the residue removed, the remaining liquid was re-centrifuged at 5000 rpm (Boeco, C-28A) for 1 h, and the supernatant, containing the exfoliated nano-sheet MoS$_2$ was collected.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

2D layered materials, aggregation-induced emission luminogens (AlEgen), exfoliation, hydrostatic strain

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