Type-I Heterostructure and Improved Phase Stability of Formamidinium Lead Iodide Perovskite Grown on WS₂

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Type-I Heterostructure and Improved Phase Stability of Formamidinium Lead Iodide Perovskite Grown on WS2

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Abstract: Heterostructures of two-dimensional materials provide an opportunity for the discovery of novel properties and improved material performances. In this work, we demonstrate the chemical vapor deposition growth of two-dimensional perovskite/WS2 heterostructures. Owing to type-I band alignment, photoluminescence (PL) measurements indicated the photogenerated carrier transfer from the WS2 to the perovskite layer. In order to investigate the carrier transfer mechanism, power-dependent PL spectra were measured. The PL intensity of the two-dimensional perovskite grown on WS2 exhibited super-linear dependence to the excitation power. In addition, two-dimensional perovskites grown on WS2 showed better air stability compared with the pristine perovskite. The improved stability is attributed to strain in the two-dimensional perovskite layer induced by the underlying WS2. The results presented here offer better understanding of the two-dimensional perovskite based heterostructures and their potential use in practical applications, such as light emitting diodes and lasers.

Keywords: two-dimensional materials; hybrid halide perovskites; heterostructure, WS2

1. Introduction

Low-dimensional materials, such as nanoparticles, nanotubes, and nanosheets, offer great potential compared to their bulk counterparts⁴⁻⁵. In particular, two-dimensional (2D) materials have been extensively studied due to their unique and diverse properties⁶⁻⁷. The most well-known 2D materials are graphene, transition metal dichalcogenides (TMDCs), and hexagonal boron nitride (h-BN)⁷. Recently, 2D hybrid halide perovskites have been reported as a new addition to the 2D materials family⁸. 2D hybrid halide perovskites are a subclass of the hybrid halide perovskite family, which have a chemical formula ABX3, where A is an organic amine cation, B is a metal cation, and X is a halide anion⁹. Within a decade, these perovskites have shown promising applications in solar cells¹⁰, photodetectors¹¹, and light emitting diodes (LEDs)¹². Compared to their bulk counterparts, 2D hybrid halide perovskites have several advantages, such as better stability, bandgap tuning by thickness engineering, and higher exciton binding energy⁸. In addition, 2D perovskites exhibit novel properties due to strong quantum confinement effect¹³.

Van der Waals heterostructures of 2D materials offer new phenomena in next generation device applications, such as interlayer excitons, valleytronics, and moiré potentials¹⁴⁻¹⁶. Recently, heterostructures of 2D perovskite with other 2D materials have been reported. These heterostructures exhibited improved performances in device applications and novel properties. Very recently, we demonstrated a perovskite/TMDC heterostructure with type-II band alignment¹⁷. In type-II heterostructures, electrons and holes are separated into different layers due to the staggered alignment of valance and conduction bands of each layer. Type-II heterostructures of 2D materials have been widely studied for charge separation, which is an important phenomenon for solar cells¹⁸. On the other hand, in type-I band alignment electrons and holes are confined in the small bandgap material, which are transferred from the large bandgap material. These confined charge carriers (electrons and holes) in heterostructures with type-I band alignment offer a great potential in optoelectronic applications, such as LEDs¹⁹. However, type-I heterostructures based on 2D materials have been scarce.

In this work, we report a 2D perovskite/tungsten disulfide (WS2) heterostructure with type-I band alignment. We employed a small bandgap 2D perovskite, namely, formamidinium lead iodide (FAPbI3). FAPbI3/WS2 heterostructures were fabricated by chemical vapor deposition (CVD) methods. The photoluminescence (PL) measurement of the FAPbI3/WS2 heterostructure revealed a PL enhancement in the FAPbI3 emission, while WS2 peak showed PL
quenching. This is attributed to the transfer of photogenerated carriers from the WS2 layer to FAPbI3 because of type-I band alignment. Power dependent PL measurement also supported the transfer of both electrons and holes from WS2 to FAPbI3. In addition, the FAPbI3 grown on WS2 showed higher stability over long time compared to FAPbI3 only sample. We discuss the origin of this improved stability in detail.

2. Experimental

2.1 CVD growth of FAPbI3/WS2

First, monolayer WS2 was synthesized by CVD on c-plane sapphire, as reported previously from our group. WO3 and sulfur powders were placed in a three-zone furnace. The WO3, sulfur, and substrates were heated at 1065 °C, 165 °C, and 925 °C, respectively. This as-grown WS2/sapphire substrate was subjected to the following two-step process to synthesize FAPbI3 perovskite layer on the WS2 surface. First step was the vapor-deposition of PbI2 on WS2. For this, PbI2 powder was placed at the center of a CVD furnace, and as-grown WS2/sapphire substrate was put at the downstream. While flowing 50 ccm of Ar gas, the furnace temperature was gradually increased to 360 °C and kept at this temperature for 15 min to deposit PbI2 on WS2. PbI2 was then converted to perovskite (FAPbI3) by intercalating formamidinium iodide at 150 °C for 20 min under a constant Ar flow.

2.2 Characterizations

Optical images were taken by an optical microscope (Nikon Eclipse ME600). Atomic force microscope (AFM) images were obtained using a Nanoscope V (Bruker). PL spectra were collected with a Raman spectroscope (Tokyo Instruments Nanofinder 30) using a 532 nm laser. UV-vis absorption spectra were measured by using a UV-3600 (Shimadzu) spectrophotometer.

3. Results and discussion

Figure 1a and b shows the optical images of monolayer WS2 before and after the deposition of a 2D FAPbI3 layer, respectively. As-grown WS2 grains have triangular shapes, with some of the grains merging with each other. White contrast seen at the center of some grains is multilayer WS2 which acted as nucleation cite. As can be seen in Fig. 1b, FAPbI3 perovskite layer selectively grew on the WS2. Previously, we reported this unique selective growth behavior for MAPbI3 perovskite/WS2 heterostructures. Our analyses revealed that the higher surface energy of
WS₂ compared to c-plane sapphire assists the growth of 2D perovskite layer₁⁷. As the selective growth takes place during the deposition of the PbI₂ layer, we observe a similar growth behavior in FAPbI₃. Here, the higher surface energy of WS₂ compensates the energy barrier required for the nucleation of PbI₂. In Fig. 1b, the red and yellow colored areas correspond to thick perovskite layers that grew around the nucleation centers of WS₂ grains. The AFM images of the WS₂ and FAPbI₃/WS₂ heterostructure are shown in Fig. 1c and d, respectively. Pristine WS₂ has a thickness around 0.9 nm, which agrees well with the reported value₂¹. In the case of the heterostructure (Fig. 1d), total thickness increased to ~5 nm, indicating that FAPbI₃ thickness is around 4 nm. These results confirm the deposition of a 2D thin layer of FAPbI₃ on WS₂. To further verify the formation of the heterostructure, PL and absorption spectra were also measured (see Fig. 1e). PL spectrum of the heterostructure (red data in Fig. 1e) consists of two distinct PL peaks. The PL peak at 1.61 eV is originated in the 2D FAPbI₃, while the peak at 2 eV is the emission from WS₂ layer₂⁰. For bulk FAPbI₃ perovskites optical bandgap is around 1.55 eV₂². However, in our case the bandgap increased to 1.61 eV as can be seen in the blue shifted PL emission. This blue shift can be explained by the quantum confinement effect in 2D FAPbI₃ due to the reduced thickness₁₇,₂³. Absorption spectrum of the heterostructure (blue data in Fig. 1e) agrees well with the PL spectrum. A sharp absorption edge at ~1.58 eV arises from FAPbI₃, while additional absorption edge at ~2 eV is from the WS₂ layer.

To further elaborate the optical properties, PL spectra of isolated FAPbI₃ and WS₂ were compared with that of the heterostructure in Fig. 2a. As expected, the pristine FAPbI₃ and the pristine WS₂ showed PL emission at 1.58 eV and 2 eV, respectively. Interestingly, in the case of the heterostructure, the PL intensity of FAPbI₃ considerably increased. In contrast, the PL from WS₂ was strongly quenched. According to the band structure calculations, the heterostructure of FAPbI₃ and WS₂ possesses type-I band alignment₁⁷,₂₄-₂₆. The type-I band alignment is displayed in Fig. 2b. In this type-I alignment, photogenerated charge carriers (both electrons and holes) transfer from the large bandgap material (WS₂) to the small bandgap material (FAPbI₃). Therefore, the increased PL intensity of FAPbI₃ in the heterostructure can be explained by energy transfer from the WS₂ layer to the FAPbI₃ layer upon photoexcitation₁⁹. PL peak intensity mappings for WS₂ peak measured before and after FAPbI₃ deposition are shown in Fig. 2c and d, respectively. It is clear that after the FAPbI₃ deposition, the PL intensity of the WS₂ decreased on the whole grain. We note that deposition of another material can cause a decrease in the PL intensity by preventing the laser penetration. However, considering the optical absorption coefficient of FAPbI₃ (up to 10⁵ cm⁻¹)²⁷, penetration depth for a 532 nm laser is around 100 nm. Therefore, we excluded the screening effect by FAPbI₃ on the PL quenching of WS₂ peak.

In order to understand the increased PL intensity of FAPbI₃ in the heterostructure, we measured excitation power dependence of the PL spectrum. In Fig. 3,
integrated PL intensity of the FAPbI3 peak is plotted as a function of the laser power. Fig. 3 displays the power dependence of the PL intensity measured for FAPbI3 only peak (black dots) and FAPbI3 peak in the heterostructures (red dots). The data can be fitted by a power-law function ($I \propto P^k$), where $I$ is the PL intensity, $P$ is the excitation power, and $k$ is the power factor. Depending on the charge carrier recombination mechanism, different $k$ values can be observed. $k < 1$ is indicative of free-to-bound transitions or Auger process, while $1 < k < 2$ is for excitonic or biexcitonic transitions. Power factors were calculated based on power-law function fittings shown by yellow lines in Fig. 3. For the perovskite only peak, $k = 0.79$ refers to the recombination of free carrier or Auger recombination. For the FAPbI3 peak in the heterostructure, a super-linear power dependence with $k = 1.32$ was observed. This super-linear dependence can be related to recombination of mixed free carrier and excitonic states. This sublinear ($k = 0.79$) to super-linear ($k = 1.32$) transition can be attributed to the disassociation of excitonic species at the FAPbI3/WS2 interface as a result of carrier transfer in type-I band alignment. However, the charge carrier dynamics of the FAPbI3/WS2 heterostructure need to be further investigated.

Finally, long-term PL stability of the heterostructure was also investigated. In Fig. 4a and b, PL spectra of FAPbI3 only and FAPbI3 grown on WS2 were measured for up to two weeks. In both cases, the PL intensity decreased with increasing the time. It is known that FAPbI3 undergoes a phase transformation at room temperature. $\alpha$-phase of FAPbI3 is the desired perovskite structure. However, $\alpha$-FAPbI3 is unstable at room temperature and it gradually converts to non-perovskite $\delta$-phase structure. The $\delta$-phase of FAPbI3 is undesirable as it does not show PL emission. Therefore, the decrease in the PL intensity is a sign of phase transformation of FAPbI3 into non-perovskite $\delta$-phase. In Fig. 4c, decrease in the PL intensity of FAPbI3 only (black line) and FAPbI3 grown on WS2 (red line) is compared. In the case of FAPbI3 only sample, PL intensity decreased almost 85% with respect to the initial intensity. For FAPbI3 grown on WS2, PL intensity only decreased to ~67%. The better stability of the latter can be attributed to the slower phase transformation of $\alpha$-FAPbI3 into $\delta$-FAPbI3. It is likely that WS2 layer suppresses the structural transformation of FAPbI3. Very recently, Chen et al., reported the phase stabilization of FAPbI3 through strain engineering. They claimed that strain is induced by heteroepitaxial growth of FAPbI3 on lattice mismatched substrates, which stabilizes the $\alpha$-FAPbI3 structure. Previously we also reported the epitaxial growth of 2D perovskite on WS2. Therefore, most likely WS2 may induce a strain on FAPbI3, which stabilizes the $\alpha$-phase by preventing the phase transformation. In Fig. 4d, an atomic model of FAPbI3/WS2 heterostructure is shown. The $\alpha$-FAPbI3 has a larger lattice constant (6.35 Å) compared to WS2 (3.15 Å). Therefore, this large lattice mismatch is expected to cause a compressive strain on the $\alpha$-FAPbI3 lattice. As conversion to $\delta$-FAPbI3 is triggered by the internal tensile strain, the compressive strain caused by WS2 cancels out the internal tensile strain in $\alpha$-FAPbI3.
4. Conclusions

We present the CVD growth of a 2D heterostructure of FAPbI$_3$ and WS$_2$. Type-I band alignment in the heterostructure enhanced the PL emission from the FAPbI$_3$ layer as a result of photogenerated energy transfer from the monolayer WS$_2$ to the FAPbI$_3$ layer. The power dependent PL measurements for FAPbI$_3$ in the heterostructure showed a super-linear dependence, which indicates the excitonic recombination at the FAPbI$_3$/WS$_2$ interface. These results hold a promise for realizing type-I device structure based on 2D perovskites. Type-I heterostructures are promising in LED and laser applications due to the enhanced light-matter interactions because of the confined charge carriers. Finally, the heterostructure showed an improved phase stability of α-FAPbI$_3$, likely due to WS$_2$-induced strain. As the long-term stability of perovskites is a major concern, we believe that our result would also contribute to the stabilization studies of perovskite structures.

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