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N-type Doping Effect of Transferred MoS_2 and WSe_2 Monolayer

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Abstract

We investigated CVD-synthesized MoS_2 and WSe_2 monolayers before and after the transfer process. N-type doping effects for both TMDs after transfer were observed by PL, Raman and conductance measurements. The effect is attributed to defect generation induced by the mechanical release from the substrate, which was also confirmed by PL measurements at low temperature.

1. Introduction

Large-area two-dimensional (2D) transition metal dichalcogenides (TMDs) synthesized by chemical vapor deposition (CVD) have been widely investigated. However, the choice of substrates are often limited by the high growth temperature (typically above 700℃) used for CVD synthesis. Numerous methods have been developed in order to transfer CVD-synthesized TMDs to arbitrary substrates [1], which is critical to enable future TMD-based flexible electronics [2] and heterogeneous integration with other materials. However, how the transfer process affects the properties of TMD films has not been comprehensively investigated. In this study, we report on an n-type doping effect of CVD synthesized MoS_2 and WSe_2 after transfer and examine its root cause.

2. Experiment Procedures

MoS_2 and WSe_2 were synthesized on sapphire substrates using the CVD technique. MoS_2 was deposited by sulfurizing MoO_3 (at 700℃ with Ar carried gas), while WSe_2 was deposited by selenizing WO_3 (at 850℃ with Ar/H_2 carried gas) powders (Fig. 1). The as-deposited MoS_2 and WSe_2 were n-type and p-type conducting, respectively, confirmed by field-effect measurements. After synthesis, MoS_2 and WSe_2 were transferred by a surface-energy-assisted process (Fig. 2) [1]. Polymethylmethacrylate (PMMA) was first coated on MoS_2 & WSe_2, then immersed into ammonia and BOE, respectively [3-4]. TMD films were released from sapphire because of the significant difference in surface energy between TMDs and sapphire. Optical properties before and after TMD transfer were characterized by photoluminescence (PL) and Raman spectroscopy. Low temperature PL (LTPL) at 4K was also performed for defect characterization.

3. Results and Discussion

Fig. 3 and Fig. 4 show the PL and Raman spectroscopy of MoS_2 and WSe_2 measured before and after transfer. Both samples were transferred to sapphire in order to eliminate the substrate effect. For MoS_2, decreased PL intensity with an apparent red-shift of the peak was observed after transfer (Fig. 3 (a)). Raman spectroscopy (Fig. 3 (b)) shows about 1 cm^{-1} shift of the A_{1g} phonon mode. Both PL and Raman spectroscopy confirm that transferred MoS_2 became more n-type doped [5-6]. However, PL and Raman spectroscopy of as-deposited samples (without PMMA coating and transfer) immersed into the ammonia solution were intact, indicating that the doping effect was not induced by the wet etching (Fig. 3 (c-d)). The optical properties shown in Fig. 4 also confirm a similar n-type doping effect in WSe_2 after transfer [7]. One plausible explanation is that this n-type doping effect was induced by the generation of S/Se vacancies [8] during the mechanical release process. This effect is favorable for n-type MoS_2 but undesirable for p-type WSe_2. Fig. 5 shows the conductance measurements of the pristine and transferred films. The conductance increased in transferred MoS_2 (with Mo/Ni contact) but decreased in WSe_2 (with Pd contact). LTPL (Fig. 6) taken at 4K shows strong defect-bound exciton (X_B) peaks induced by the transfer process [9], confirming the defect formation.

4. Conclusions

The n-type doping effect for both MoS_2 and WSe_2 has been confirmed by PL, Raman, and conductance measurements. This effect was not induced the wet chemical etching, but more likely by the defect generation during the mechanical release process. S/Se vacancies are known to introduce n-type doping in TMDs, and significant defect-bound exciton peaks are also observed at low temperature. These findings suggest that the transfer process can be engineered to enhance the n-type doping and thus device performance of MoS_2. However, further optimizations are required to minimize its adverse impact on p-type WSe_2.

References

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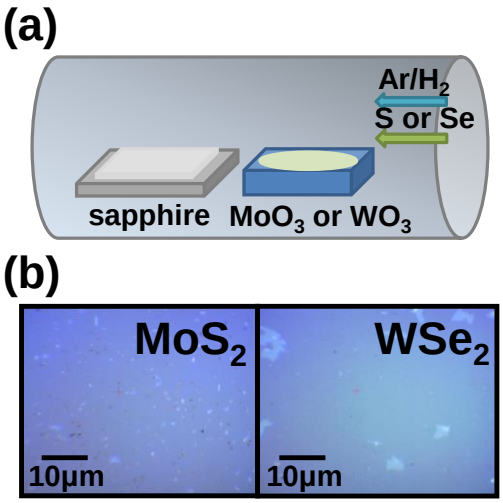


Fig.1 (a) Schematic illustration of CVD synthesis of TMDs in a furnace. (b) Optical image of the as-deposited MoS_2 and WSe_2 monolayers on sapphire substrates.

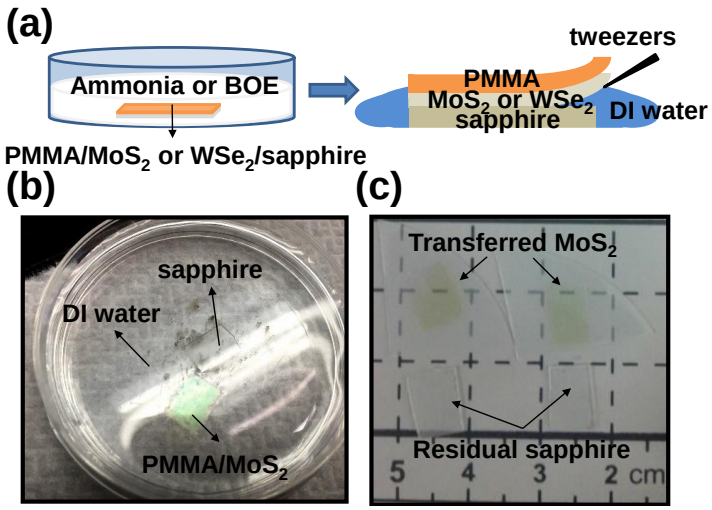


Fig.2 (a) Schematic diagram of the transfer process for MoS_2 and WSe_2. (b) PMMA/TMDs were released from sapphire assisted by different surface energy between TMDs and sapphire. (c) Transferred MoS_2 and residual sapphire substrates.

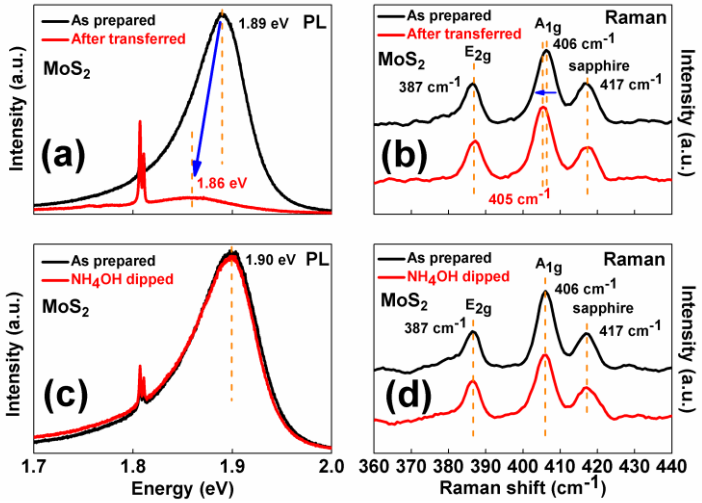


Fig.3 (a-b) PL and Raman spectroscopy of MoS_2 before and after transfer. PL intensity was severely degraded with an apparent red shift, and A_{1g} phonon mode was also shifted. The optical measurements indicate the n-type doping after transfer. (c-d) Optical properties of the as-deposited MoS_2 films before and after immersed into ammonia, showing no apparent difference.

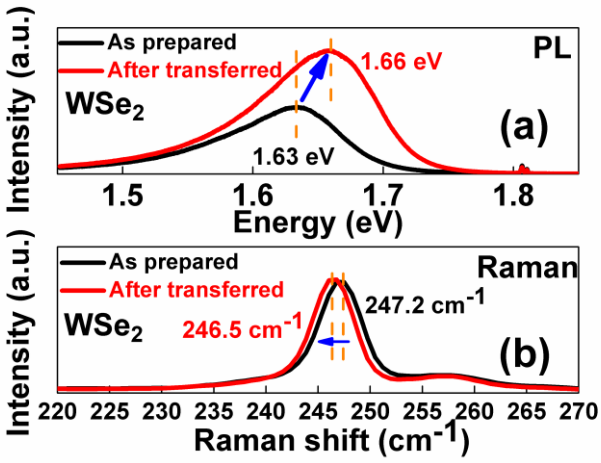
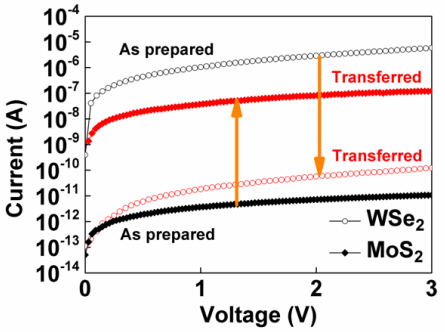


Fig.4 (a) PL and (b) Raman spectroscopy of WSe_2 before and after transfer. PL intensity was increased with an apparent blue shift, and Raman peak was also shifted.



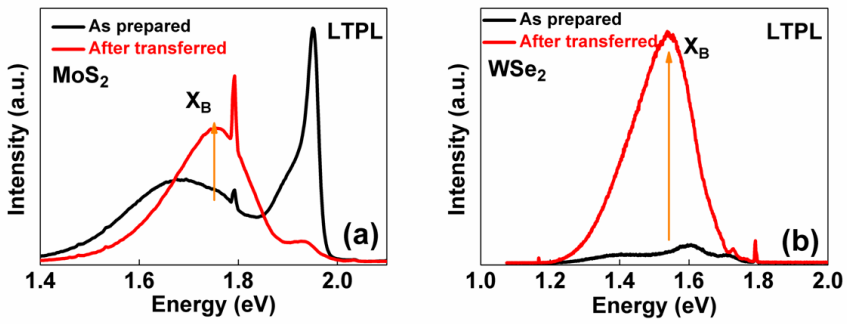


Fig.5 Conductance measurements of MoS_2 and WSe_2 before and after transfer. The conductance was increased for n-type MoS_2 while decreased for p-type WSe_2 because of the n-type doping effect.

Fig.6 LTPL measurements at 4K to characterize defects of pristine and transferred (a) MoS_2 and (b) WSe_2 films. X_B peak was significantly increased because of the neutral excitons bound to the defects induced by the transfer process.