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Oxygen-Assisted Nucleation Control for High-Quality Metalorganic Chemical Vapor Deposition of Transition Metal Dichalcogenides

As compared with conventional powder-based chemical vapor deposition (CVD) of two-dimensional transition metal dichalcogenides (TMDCs), metalorganic chemical vapor deposition (MOCVD) has its advantages in terms of scalability and uniformity. Since the growth of atomically-thin TMDCs by MOCVD was first reported in 2015, [1,2] several groups have also demonstrated to grow MOCVD-TMDCs with the same or similar precursors and growth conditions. However, up until now, most of MOCVD-TMDCs has its crystal size or grain boundary period of sub-micron scale, which became a primary scattering source to limit the mobility of TMDCs when they are used as atomically thin channels.

Generally, MOCVD of TMDCs involve H_2 gas injection during the reaction to assist the chemical decomposition of chalcogen precursor (ex. $(C_2H_5)_2S$) at the reaction temperature. [1,2] Also, during the growth, the level of reactant gases is maintained as constant, which makes it hard to control nucleation and growth process independently. Add to that, the lack of understandings on the growth mechanism hindered further engineering on the quality.

To solve the problem, we suggested a flow of O_2 gas during the MOCVD growth. It is likely that O_2 participated in the decomposition of $(C_2H_5)_2S$ precursor as well as in the formation of oxysulfide nuclei (Mo_xO_ySz) at the initial stage of the growth. Interestingly, when the supply of O_2 was cut off at peak temperature, the crystallinity of TMDCs were greatly improved. The average crystal size was ~100 folds larger (few tens of μm) compared with the previous reports. It was found that the fast exhaustion kinetics of O_2 gas by rapid reactions accompanied by O_2 played key roles in nucleation density control, as supported by growth system simulations. Also, with the suggested method, growth time was significantly decreased to <1 hour, which implies economic production of MOCVD-TMDCs in a batch scale for future electronics and optoelectronics.

References

[1] Kibum Kang, Saien Xie, Lujie Huang, Yimo Han, Pinshane Y. Huang, Kin Fai Mak, Cheol-Joo Kim, David Muller and Jiwoong Park, Nature, 520 (2015) 656-660
[2] Sarah M. Eichfeld, Lorraine Hossain, Yu-Chuan Lin, Aleksander F. Piasecki, Benjamin Kupp, A. Glen Birdwell, Robert A. Burke, Ning Lu, Xin Peng, Jie Li, Angelica Azcatl, Stephen McDonnell, Robert M. Wallace, Moon J. Kim, Theresa S. Mayer, Joan M. Redwing and Joshua A. Robinson, ACS Nano, 9 (2015) 2080-2087

Figures

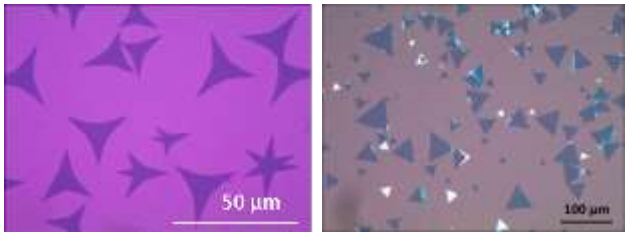

Figure 1: Optical microscope images of MOCVD-MoS_2 and WS_2