Atomic-scale vacancy healing mechanism in monolayer MoS_2 via chemical treatment

Choi, W.[1], Kim, D.[1], Han, G.[1] and Kim, Y.[2]

[1] Department of Energy Science, Sungkyunkwan University, Republic of Korea, [2] Department of Energy Science, Sungkyunkwan University (SKKU), Republic of Korea

Monolayer transition metal dichalcogenides (1L-TMDs) are promising two-dimensional semiconductors with direct bandgap, suitable for atomically thin nanophotonic device applications. However, the very low quantum yield of intrinsic 1L-TMDs due to a high density of chalcogen vacancies is a major drawback for practical use, which has thus drawn extensive studies on enhancement of light emission from 1L-TMDs. Among them, chemical treatment using bis(trifluoromethane) sulfonimide (TFSI) has been revealed to be particularly effective to increase the photoluminescence (PL) of 1L-MoS_2. However, the underlying atomic mechanism of PL enhancement has remained elusive, which is fundamental knowledge to tailoring optoelectronic properties of 1L-TMDs via facile chemical approaches. Here, we report microscopic origin of defect healing observed in TFSI-treated 1L-MoS_2 through a correlative combination of optical characterization and atomic-scale scanning transmission electron microscopy (STEM) combined with DFT calculations. We found that more than 90% of sulfur vacancies can be directly repaired by extrinsic sulfur atoms sourced from the dissociation of TFSI, concurrently resulting in the significant PL enhancement. In the course of vacancy healing, reactive sulfur dioxide molecule dissociated from TFSI can be reduced to sulfur ion and oxygen gas at the vacancy site to form a strong bonding of S-Mo, which is energetically favorable. Our results underline how defect-mediated nonradiative recombination can be effectively removed by simple chemical treatment on the atomic level.

[1] We thank S. Roy and Prof. J. Kim in Sungkyunkwan University for the chemical treatment and optical measurements for the samples and Dr. H. Kim and Dr. S.J. Yun in the same university for the CVD grown MoS_2 samples and S. Jeon and Prof. J. Lee in Pusan National University for the DFT calculations.

[2] This work was supported by the Brain Korea 21 Plus program and the Institute for Basic Science (IBS-R011-D1) and Creative Materials Discovery Program through the NRF (National Research Foundation of Korea) grant (NRF-2015M3D1A1070672) in Korea.