Observation of the annealing-induced phase-transition in monolayer-MoS_2 on gold film

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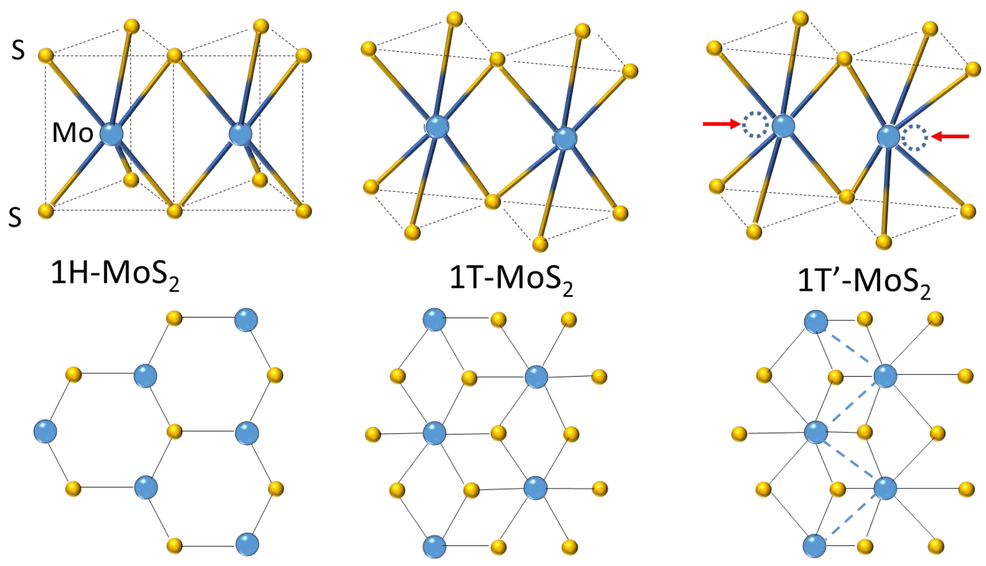
Abstract

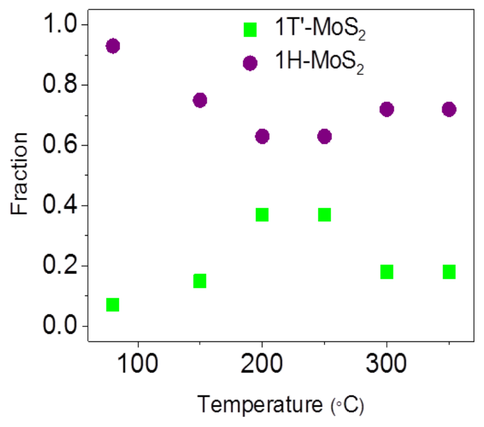
The polymorphic features of two-dimensional transition metal dichalcogenides materials such as Molybdenum Disulfide (MoS_2) exhibit unique and fascinating optical and transport properties with immense potential in device applications. Here, we report the structural phase transition of monolayer MoS_2 from the trigonal semiconducting 1H-phase to the distorted octahedral quasi-metallic 1T'-phase. Of which, we observe a tunable inverted gap (~0.50eV) and a fundamental gap (~0.10eV) in quasi-metallic monolayer-MoS_2. Using spectral-weight transfer analysis, we find that the inverted gap is attributed to the strong charge-lattice coupling in 2D-TMDs. Using a comprehensive experimental study involving transport, Raman, photoluminescence (PL) and synchrotron-based photoemission spectroscopy (PES), supported by theoretical calculations, we monitor the 1H-to-1T' phase-transition in monolayer-MoS_2/Au as a result of high-temperature annealing and study the changes in its optical and electronic properties. We made further clarifications that the effects of electron-doping from gold, further facilitated by the presence of interfacial tensile strain, are the primary mechanisms which result in this 1H-1T' structural phase transition, thus resulting in the formation of the inverted and fundamental gaps. Results from our study highlight the pivotal role that charge-lattice coupling play that lead to the intrinsic properties of the inverted and fundamental gaps and polymorphism of MoS_2, thereby unleashing new possibilities for the use of 2D-transition metal dichalcogenide-based device fabrication.

Polymorphism of monolayer-MoS_2



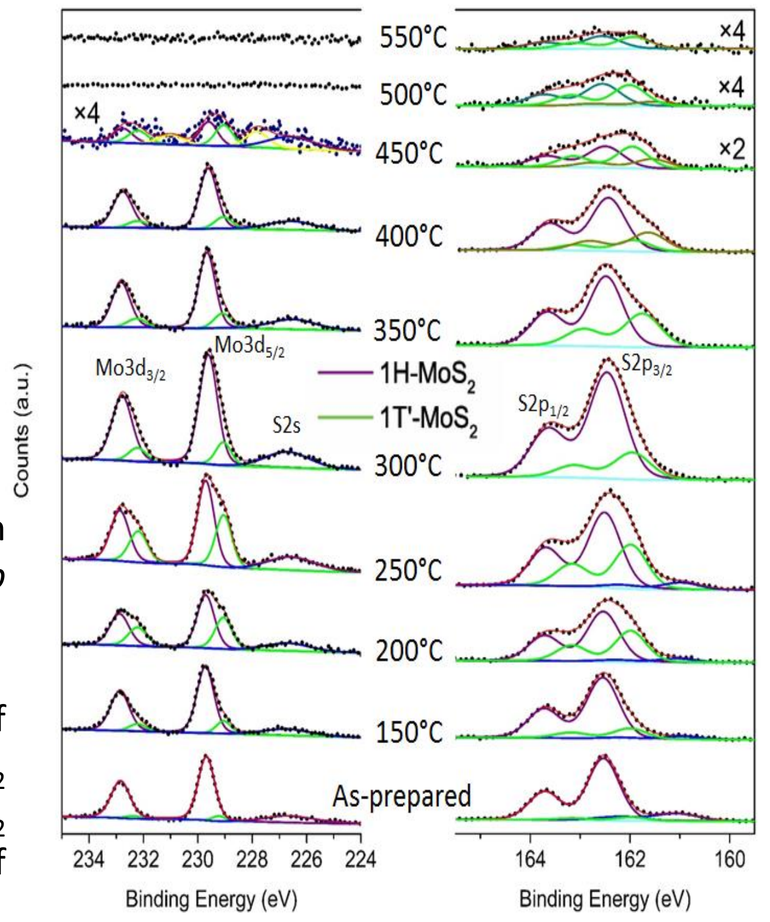
Three main phases that two-dimensional MoS_2 comprises. Trigonal semiconducting 1H-phase with a direct bandgap. Octahedral metallic 1T-phase which is reported to be unstable in free-standing conditions and will spontaneously relax to a distorted 1T' structure. Distorted transition metal atoms in the 1T'-phase form a period doubling 2×1 structure consisting 1D zigzag chains.

X-ray photoelectron spectroscopy



The core level photoemission spectra of Mo3*d* and S2*p* after annealing.

Extracted relative fraction of 1H-MoS_2 and 1T'-MoS_2 components of Mo3$d_{5/2}$ peaks as functions of temperature.



Electric and ellipsometry

Observation of the inverted gap (~0.5 eV) and fundamental gap (~0.1 eV), which are the symbols of 1T'-phase.

The electrical conductivity results show the higher conductivity for 1T'-phase MoS_2.

Raman

The red shifts of both A_{1g} and E^1_{2g} modes indicate the increase of electron transfer and tensile strain after annealing which result in the 1H-1T' phase transition of MoS_2.

The J1, J2, J3 peaks are the important evidences for 1T'-phase MoS_2.

Reference:

Xinmao Yin, Qixing Wang, Liang Cao, Chi Sin Tang, Xin Luo, Yujie Zheng, Lai Mun Wong, Shi Jie Wang, Su Ying Quek, Wenjing Zhang, Andrivo Rusydi, Andrew T.S. Wee. Tunable inverted gap in monolayer quasi-metallic MoS_2 induced by strong charge-lattice coupling. *Nature Communications* **8**, Article number: 486 (2017)