Unravelling High-Yield Phase-Transition Dynamics in Transition Metal Dichalcogenides on Metallic Substrates

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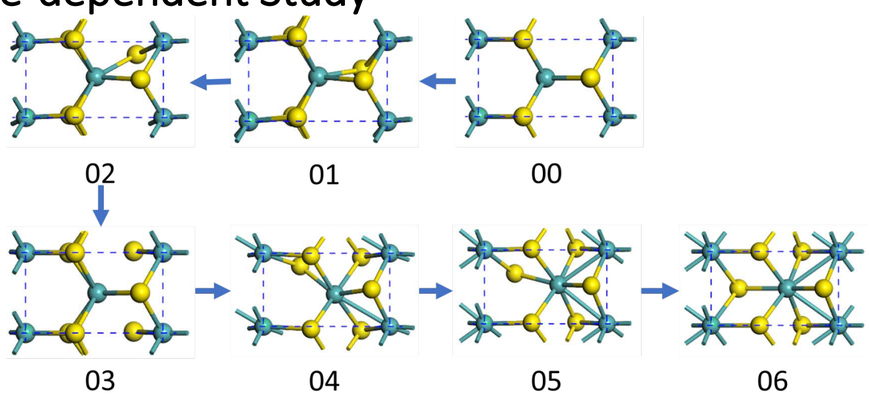
Abstract

Two-dimensional transition metal dichalcogenides (2D-TMDs) possess unique polymorphic features such as the semiconducting 1H and quasi-metallic 1T' phases withiinteresting optical and electronic properties. They are utilized in novel electronic and photonic device applications. The favorable quasi-metallic nature of 1T'-phase 2D-TMDs makes 1H-1T' phase engineering processes a vital discipline for novel device applications. We report a high-yield 1H-1T' phase transition of monolayer-MoS_2/Cu and monolayer-WSe_2/Au via an annealing-based method. Comprehensive experimental and first-principle study is performed to study the mechanism underlying the high-yield phase transition process of 2D-TMDs on metallic substrates. While each 2D-TMD possess different intrinsic 1H-1T' energy barriers, option of substrates with higher chemical reactivity is important to enhance the 1H-1T' phase transition. It is enhanced by increasing the interfacial hybridizations via increased interfacial binding energy, larger charge transfer, shorter interfacial spacing and weaker bond strength. [Adv. Sci. 2019, 6, 1802093]

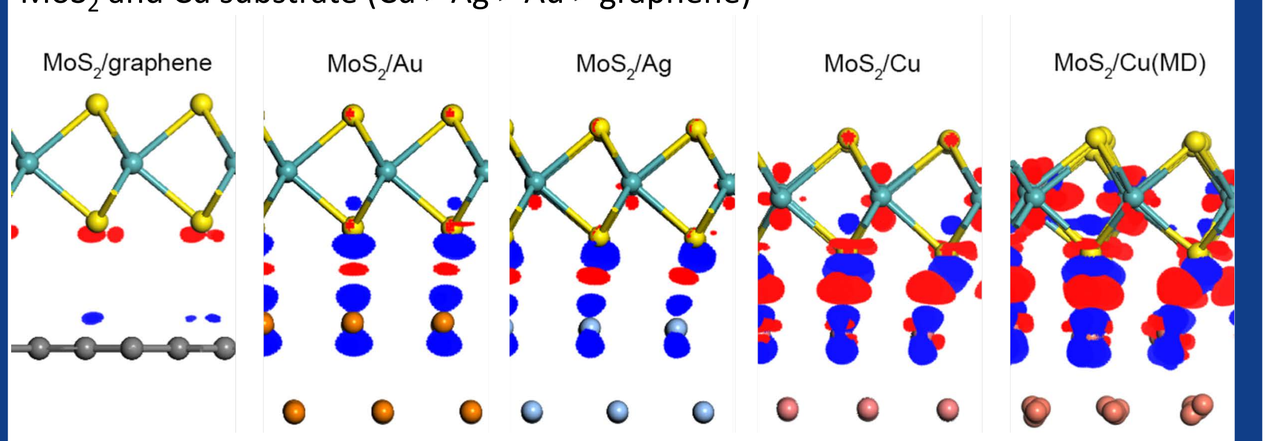
Substrate-dependent Study



Computational study comparing interfacial dynamics between 1H-phase MoS_2 on graphene, Au, Ag and Cu substrates

	Binding Energy (meV/Å²)	Charge Transfer (e⁻/f.u.)	Interfacial Distance (Å)
MoS_2/graphene	-20.5	0.005	3.38
MoS_2/Au	-55.7	0.017	2.68
MoS_2/Ag	-62.5	0.071	2.50
MoS_2/Cu	-90.8	0.126	2.16
MoS_2/Cu(MD)	-93.3	0.154	1.78

Appreciably stronger interfacial hybridization between 1H-phase monolayer-MoS_2 and Cu substrate (Cu > Ag > Au > graphene)

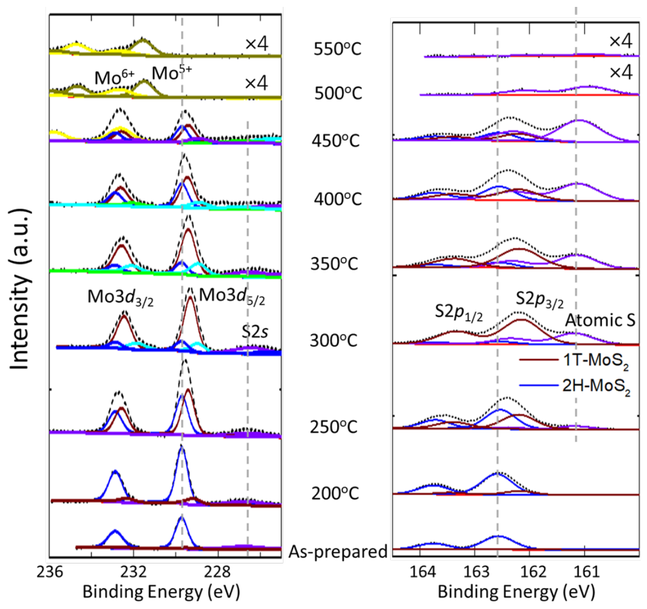


Interfacial hybridization of TMD/substrate systems increases with increasing chemical reactivity of the metallic substrates

Ab initio molecular dynamics (MD) simulate annealing process of MoS_2/Cu system at 550K. Further enhancement in interfacial hybridization compared to pristine MoS_2/Cu system

Increased interfacial hybridization at interface induced is key mechanism leading to the 2H-1T' phase transition of 2D-TMDs

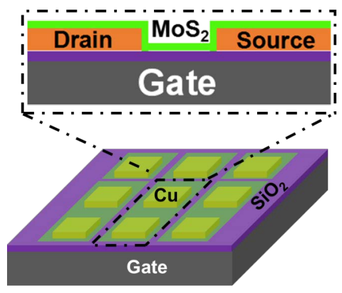
Annealing-based Phase Transition: MoS_2/Cu



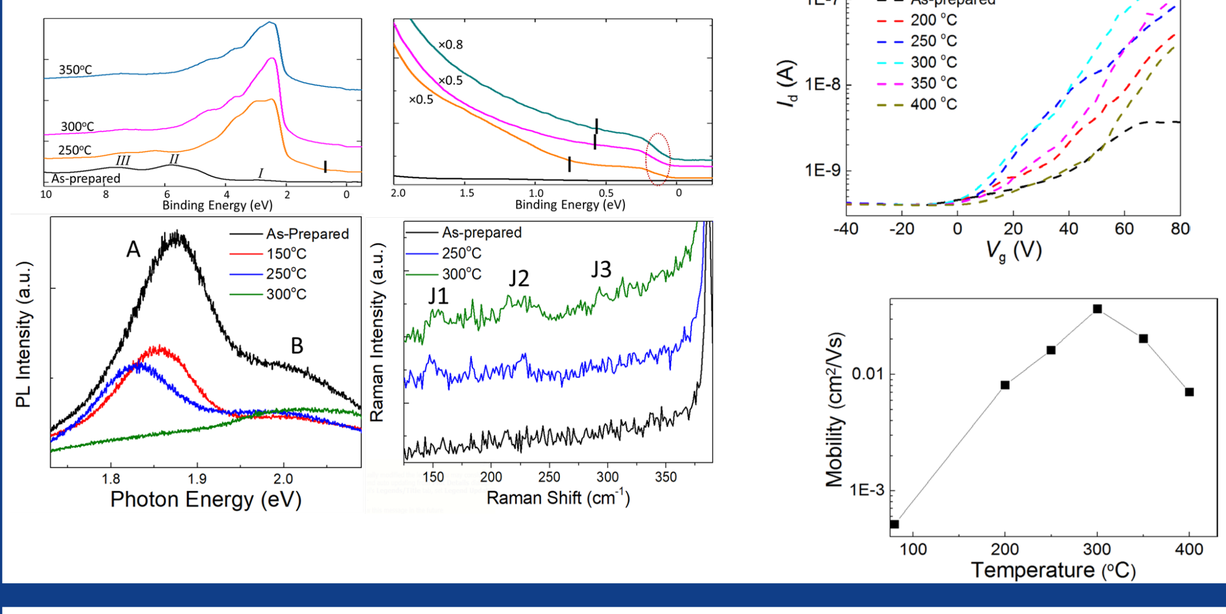
Experimental study of percentage yield in MoS_2/Cu and WSe_2/Au.

~7% 1T'-MoS_2/Cu in pristine state

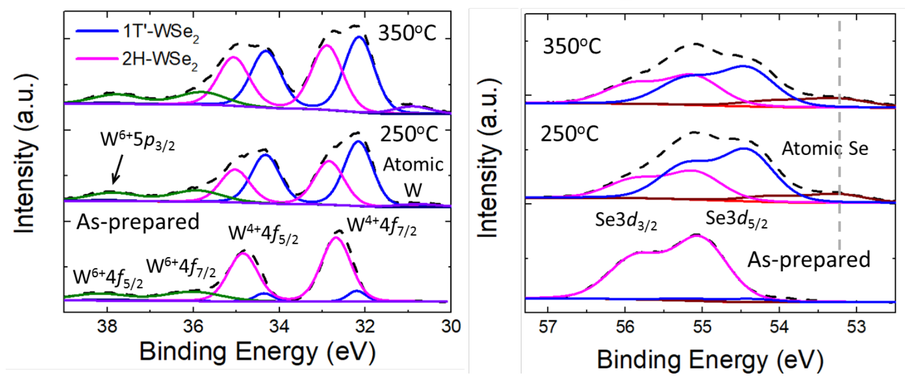
Optimum yield of ~85.7% at 300°C annealing temperature



Raman, PL, Electric Transport & UPS Data

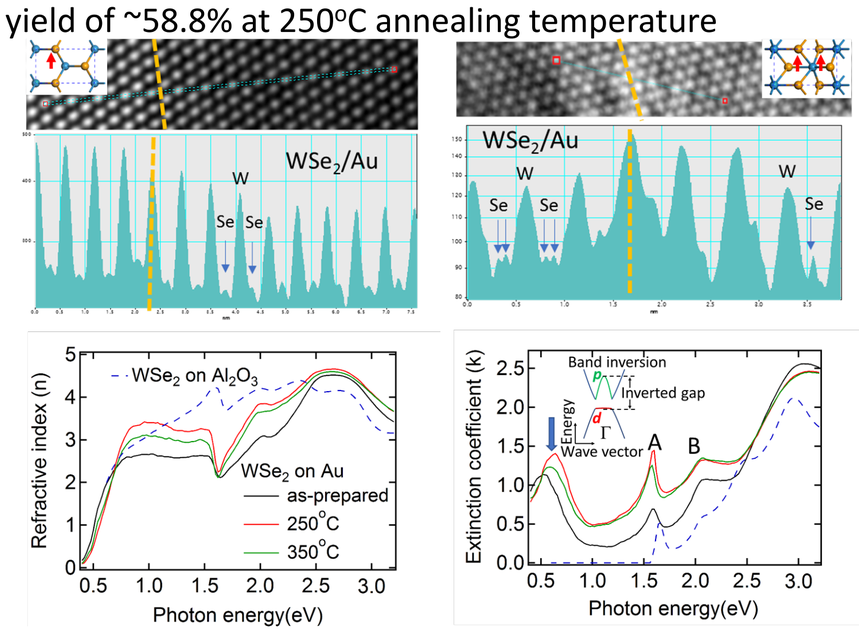


Annealing-based Phase Transition: WSe_2/Au

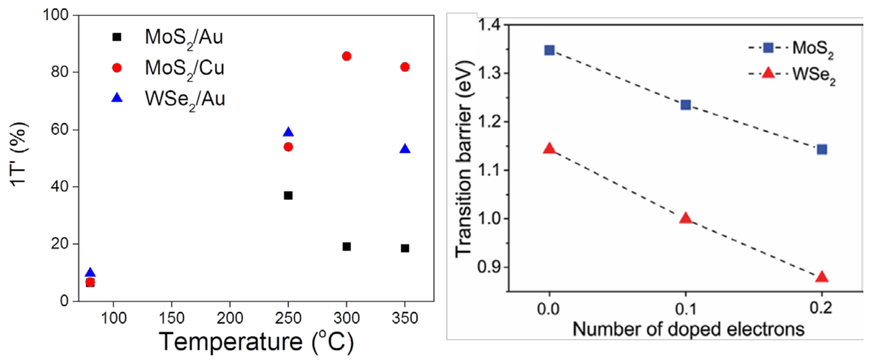


~10% 1T'-WSe_2/Au in pristine state

Optimum yield of ~58.8% at 250°C annealing temperature



Percentage 1T'-phase Yield in Different Systems



Conclusion

Collective general trends of this annealing-based phase transition study of 2D-TMDs on metallic substrates show that metallic substrate with high chemical reactivity and 2D-TMD with low intrinsic 1H-1T' energy barriers significantly dictate the yield of 1T'-phase 2D-TMD. Our study demonstrates the unprecedented quality of the 2D-TMD/metallic substrate interface at the atomic length scale. This can be a major step forward towards general interest.