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## ARPES study on monolayer CrSe<sub>2</sub> films fabricated by molecular beam epitaxy

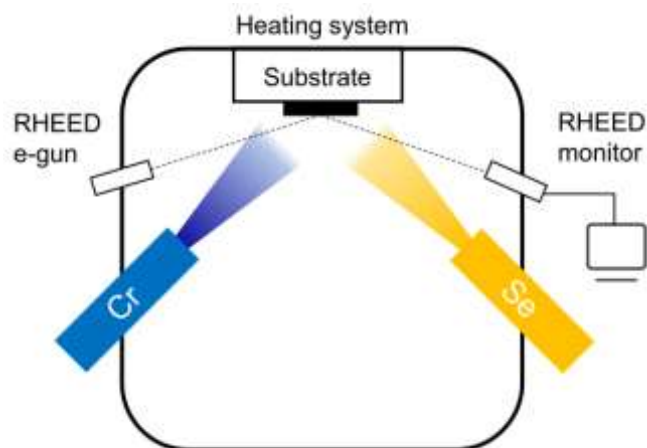
The discovery of graphene and its exotic properties paves the way for the investigations of the two-dimensional (2D) materials. Among various 2D materials, transition metal dichalcogenides (TMDCs)  $MX_2$  ( $M$  = transition metal,  $X$  = chalcogen) are ones of the ideal platforms for exploring the emergent 2D physics because they have rich combinations of constituent elements and various polytypes. So far, the group VI TMDCs monolayers such as MoS<sub>2</sub> and WTe<sub>2</sub> fabricated by utilizing the mechanical exfoliation method have been intensively investigated because of the characteristic spin-valley coupled electronic structures and topological properties. On the other hands, the state-of-the-art sample fabrication technique, molecular beam epitaxy (MBE), enables us to investigate the physics in the hardly-cleavable 3d-based TMDCs, bringing the strong electron correlation effects onto the 2D materials platform, such as the excitonic physics, charge density wave, magnetism, and so on. In fact, the emergent room temperature ferromagnetism in monolayer VSe<sub>2</sub> [1] and MnSe<sub>x</sub> [2] films grown by MBE are recently discovered, opening up the experimental investigations of exploring further 3d-based TMDC monolayers.

In this study, we focus on CrSe<sub>2</sub> as another strongly correlated 3d-based TMDCs. The previous works [3, 4] revealed that the bulk CrSe<sub>2</sub> has only 1T-phase and exhibits the successive structural and magnetic phase transitions due to the strong competitions between the multiple degrees of freedom of electrons such as the charge, atomic orbit and spin. Although the CrSe<sub>2</sub> shows intriguing physical properties, the difficulty in synthesis mainly due to the valence instability of Cr<sup>4+</sup> hinders a comprehensive understanding of the electronic structures and origins of them. Here, we report a successful fabrication of the monolayer CrSe<sub>2</sub> by MBE with our growth recipe [5]. Through the investigation on the electronic structure by utilizing the angle-resolved photoemission spectroscopy (ARPES) with the help of band calculations, the monolayer *T*-phase CrSe<sub>2</sub> is revealed to exhibit a transition to correlated insulator accompanying with the gap opening around Fermi energy. Moreover, the monolayer *H*-phase CrSe<sub>2</sub> can be stabilized by changing growth conditions and revealed to be a new valley semiconductor.

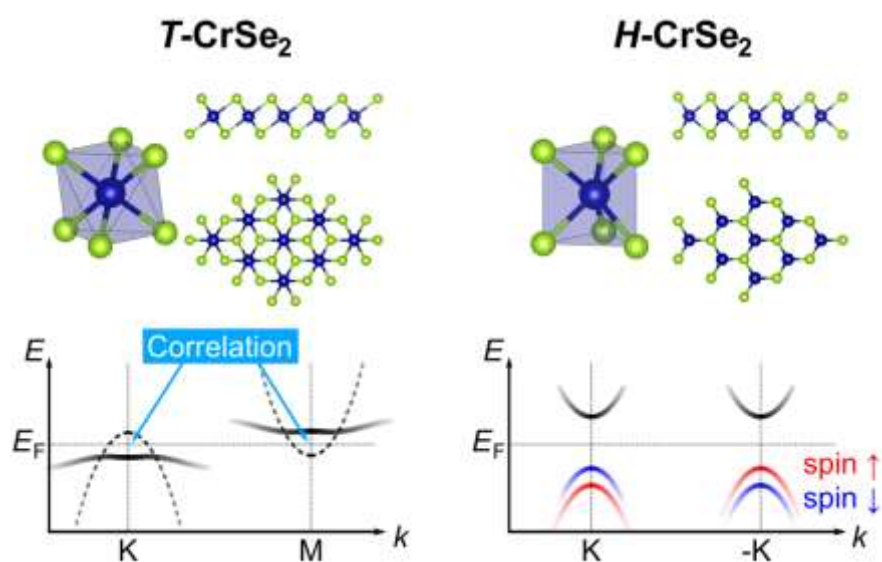
### References

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## Figures



**Figure 1:** Schematic image of our MBE equipment.



**Figure 2:** Schematic illustrations of our results on monolayer  $\text{CrSe}_2$  films.