

ARPES study of TiFe_xS_2 ($x = 0, 0.33$)

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$1T\text{-TiS}_2$ is non-magnetic layered material with $1T\text{-CdI}_2$ -type crystal structure. The hexagonal layer of Ti ions is sandwiched between the two hexagonal layers of S ions and the Ti ion is octahedrally coordinated with six S ions. The S-Ti-S triple layers are covalently bonded and these TiS_2 triple layers are weakly coupled with van der Waals (vdW) force. In the vdW gap located between the TiS_2 layers, the other $3d$ transition-metal M can be intercalated as $M_x\text{TiS}_2$. Among them, Fe_xTiS_2 exhibits a wide variety of magnetic properties [1]. With increasing the Fe concentration from $x = 0$, the cluster spin glass (CG) state with the Ising spins is found for $x < 0.20$. After exhibiting the antiferromagnetic (AFM) state up from $x = 0.20$ to 0.28 , the CG state is again realized for $0.28 < x < 0.38$ and the AFM state for $0.38 < x < 0.50$. Above $x = 0.50$, the ferrimagnetic behavior is observed. On the other hand, the Fe ion intercalated in the vdW gap between the TiS_2 layers occupy the octahedral site surrounded by six S ions. X-ray studies reveal the Fe random distribution for $x < 0.20$, while the formation of $2\sqrt{3}a \times 2a \times 2c$ or $2a \times 2a \times 2c$ superlattices due to the Fe ordering for $x = 0.25$ and $\sqrt{3}a \times \sqrt{3}a \times 2c$ superlattice for $x = 0.33$. The rich magnetic states of Fe_xTiS_2 are expected to link to the change in electronic band structure due to the Fe intercalation. In this study, we carried out angle-resolved photoemission spectroscopy (ARPES) to reveal the electronic band structure of Fe_xTiS_2 ($x = 0, 0.33$) at beamlines BL-1 and BL-7 of Hiroshima Synchrotron Radiation Center (HSRC).

Figure 1(a) shows the ARPES intensity plots of TiS_2 measured at $h\nu=66$ eV along the $\bar{\Gamma}\text{-}\bar{M}$ directions of the surface Brillouin zone, roughly corresponding to $\Gamma\text{-}M$ direction of the bulk Brillouin zone, estimated from the $h\nu$ -dependent measurements. We find an electron pocket derived from the Ti $3d$ states around the M point. Almost non-dispersive band is observed at binding energy of $E_B = 0.3$ eV, which is assigned to the $3d$ states of the Ti ions self-intercalated in the vdW gap between TiS_2 layers. The parabolic band around the Γ point is due to the S $3p$ states.

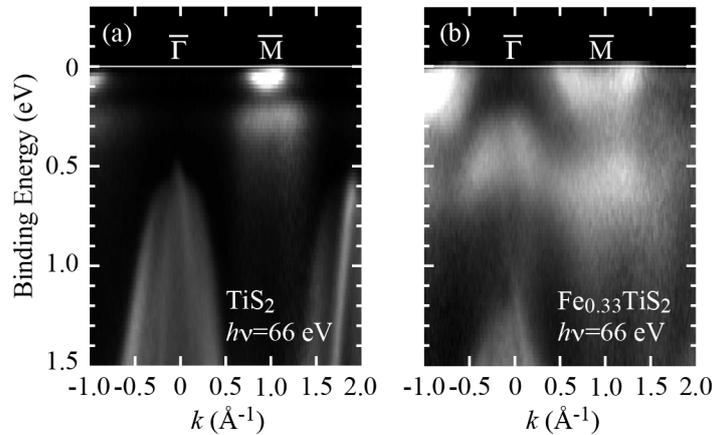


FIGURE 1. ARPES intensity plots of (a) TiS_2 and (b) $\text{Fe}_{0.33}\text{TiS}_2$ measured at $h\nu = 66$ eV along $\bar{\Gamma}\text{-}\bar{M}$ direction.

Figure 1(b) shows the ARPES intensity plots of $\text{Fe}_{0.33}\text{TiS}_2$. The electron pocket around the M point is shifted to higher binding energy. In addition, the parabolic S $3p$ band is also shifted. These results indicate that electrons are transferred from the intercalated Fe ions to host TiS_2 layers. The dispersive Fe $3d$ -derived band is observed around $E_B = 0.3 - 0.7$ eV, reflecting the Fe ions align periodically in the vdW gap parallel to the TiS_2 layers (in plane). The Fe $3d$ band seems to be bent upward before the M point, probably due to the $\sqrt{3}a \times \sqrt{3}a$ periodicity in plane for $x = 0.33$. We measured the ARPES spectra along the Γ -A direction with changing $h\nu$ from 30 to 124 eV. We find that the Fe $3d$ band exhibits the dispersion along the direction normal to the TiS_2 layers and has half period of the S $3p$ band, reflecting the $2c$ periodicity due to the formation of $\sqrt{3}a \times \sqrt{3}a \times 2c$ superlattice.

REFERENCES

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