OBSERVATION OF SPIN POLARIZATION IN TRANSITION METAL DICHALCOGENIDE

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Monolayer transition-metal dichalcogenides (TMDC) have been attracting great attention as the candidate material for next-generation electronic devices utilizing spin and/or valley degrees of freedom [1-5]. As shown in Fig. 1(a), for example, the monolayer-MoS\(_2\) is formed by the 2-dimensional network of Mo sitting in the center of S triangular prisms, thus possessing the 3-fold symmetry without an inversion center. Because of this peculiar trigonal structure, in contrast to the simple honeycomb graphene, the spontaneous spin and valley coupled band structure (i.e. spin/valley polarization) is realized at the Brillouin zone corners. Indeed, the control and detection of the valley polarization are demonstrated by using the photoluminescence circular dichroism in monolayer-MoS\(_2\) [1-4]. Monolayer-MoS\(_2\) hitherto has been mostly obtained by mechanically exfoliating the 2H-polytype bulk crystal, which itself lacks the spin/valley polarization due to its centrosymmetric structure recovered by the hexagonal stacking. The exfoliated monolayer-MoS\(_2\) thus obtained has the limited size of area (< 1 \(\mu\)m\(^2\)), thus preventing the direct investigations on its spin and electronic structure. Here we focus on another stable phase of polytype, 3R-MoS\(_2\) (space group \(R3m\)) [6-8], which is composed of the so-called \(abc\) stacking where the 3-fold symmetry of monolayer-MoS\(_2\) remains in bulk, as shown in Fig. 1(b). In 3R-MoS\(_2\), the spin/valley coupling similar to monolayer-MoS\(_2\) should reside in bulk, appearing as the opposite spin-polarizations at \(\bar{K}\)- and \(\bar{k}\)-points [Fig. 1(c)].

Spin-resolved angle-resolved photoemission spectroscopy (SARPES) with He\(\alpha\) light source (\(h\nu = 21.2\) eV) was performed by using the VLEED (Very Low Energy Electron Diffraction) spin detector at BL19A in photon factory, KEK [9]. The angular resolution was set to \(\pm 1^\circ\) and the total energy resolution was set to 100 meV. Samples (~ 1 mm \(\times\) 1 mm) were cleaved in situ near room-temperature and measured at 110 K. Measurement geometry is shown in Fig. 1(d). The VLEED detector can measure the spin polarization along \(X\) and \(Y\) directions. Note that the sample orientation \((x, y, z)\) is rotated around \(X\)-axis to measure the \(\bar{K}\)-point.
Figure 1(e) shows the SARPES spectra detecting $Y$-oriented spin ($I_Y^{\uparrow,\downarrow}$) of 3R-MoS$_2$, 2H-MoS$_2$ and 2H-WSe$_2$, measured at $\bar{k}$- and $\bar{k}'$-points. $I_Y^{\uparrow,\downarrow}$ includes the information on spin polarization along $z$ and $y$, due to the sample orientation. $I_Y^{\uparrow,\downarrow}$ data for 3R-MoS$_2$ at $\bar{k}$-point clearly indicate the huge spin polarization, i.e. spin up (down) dominant for the upper (lower) band, which gets inverted at $\bar{k}'$-point. By comparing with the ab-initio calculation, the opposite spin-polarizations at $\bar{k}$- and $\bar{k}'$- points observed in 3R-MoS$_2$ offer the evidence of the spontaneous out-of-plane spin polarization which consequently accompanies the $\bar{k}$- and $\bar{k}'$-valley polarization, as similarly expected in monolayer-MoS$_2$ [1-5]. 2H-MoS$_2$ and 2H-WSe$_2$, on the other hand, show the nearly equivalent intensities of $I_Y^\uparrow$ and $I_Y^\downarrow$, indicating that the spin polarization is absent or very weak. It should be reflecting the centrosymmetric crystal structure of 2H-polytype, which at the same time suggests the spin-splitting observed in 3R-MoS$_2$ is indeed from bulk, not arising from the inversion breaking at the surface.

To summarize, we investigated the spin- and electronic structure at the Brillouin zone corner for noncentrosymmetric 3R-MoS$_2$ and centrosymmetric 2H-MoS$_2$ and 2H-WSe$_2$ by using spin-resolved ARPES. The observed spin polarization in 3R-MoS$_2$ is identical to that expected in monolayer-MoS$_2$ and other TMDC families. The noncentrosymmetric 3R phase should be useful for future device fabrications, that will drive the valley/spin-tronics based on 2 dimensional crystals.

**REFERENCES**