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ARPES and XMCD studies of the van der Waals ferromagnet $Cr_2Ge_2Te_6$

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1 Introduction

In recent years, van der Waals (vdW) ferromagnets, which have attractive physical properties, have been intensively studied in order to investigate the origin of their ferromagnetism in 2D crystals [1]. A representative vdW ferromagnetic insulator $Cr_2Ge_2Te_6$ (CGT) exhibits ferromagnetism below ~ 61 K [2,3,4] and the magnetic moment of 2.3 μ_B/Cr atom with the easy magnetization axis along the *c*-direction [2]. As for the electric properties, resistivity measurements on CGT have shown semiconducting behaviors and its resistivity prominently increases below ~100 K [2,5]. It has also been reported that the magnetic behavior of CGT atomic layer is well described by the 2D Heisenberg model with single-ion anisotropy [6].

To unveil the origin of the ferromagnetism in 2D materials, understanding the physical properties from both the electronic and magnetic structure points of view is highly important.

2 Experiment

Single crystals of CGT were synthesized using the flux method. ARPES measurements were performed at beam line BL-28A of Photon Factory. The incident light was circularly polarized in the energy range of 34-80 eV. A SCIENTA SES 2002 photoelectron analyzer (Scienta-Omicron Co., Ltd.) was used. Measurements were performed under the vacuum of 2.0 $\times 10^{-8}$ Pa and the sample was cleaved in the analyzer chamber prior to measurements to obtain clean surfaces. The sample temperature during the measurements was set to 150 K, above the transition temperature, in order to avoid charging effects. The binding energy (E_B) was corrected by measuring the Fermi level (E_F) of an Au foil that is in electrical contact with the sample. The energy resolution was set to ~50 meV.

X-ray magnetic circular dichroism (XMCD) measurement was conducted at BL-16 of Photon Factory using a vector-type magnet. The sample temperature was set to 50 K, below the transition temperature. Magnetic field of 1 T was applied both parallel and perpendicular to the ab-plane of the sample.

3 Results and Discussion

Fig. 1 shows ARPES results. A constant energy mapping at E - E_F = -0.35 eV is shown in Fig.1(a). Strong intensity at the Γ point is observed, and the ARPES spectra taken along the red line in Fig.1(a) is shown in Fig. 1(b). Hole-like band around the Γ point below Fermi level is consistent with its insulating property.

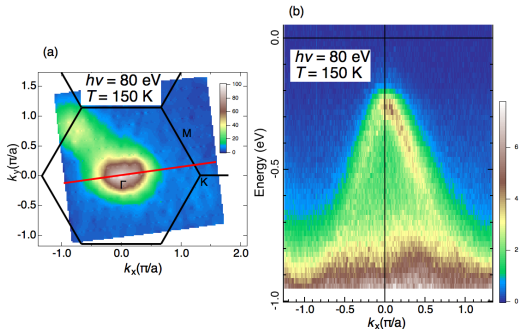

Fig. 1 ARPES results of CGT. (a) Constant energy mapping at E - E_F = -0.35 eV. (b) Hole-like band around the Γ point.

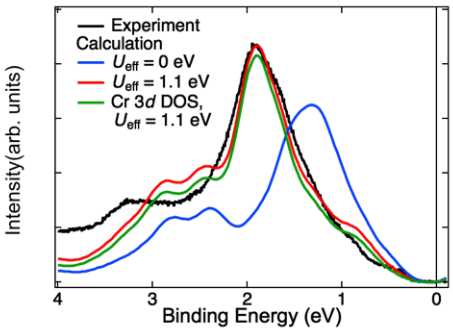

Fig. 2 *k*-integrated spectrum compared with spectra simulated by DFT calculation taking into account the photo-ionization cross-section of each atomic orbital.

We have also performed DFT calculations. The *k*-integrated spectra obtained by experiment and the calculated spectra are shown in Fig. 2. The peak at the binding energy of ~1.9 eV is not reproduced by calculation without Coulomb interaction. On the other hand, calculation including Coulomb interaction U_{eff} = 1.1 eV within the DFT+*U* method reproduces the peak position. Indeed, the value U_{eff} = 1.1 eV can reproduce the ferromagnetism in the DFT calculation, indicating that Coulomb interaction between the Cr 3*d* electrons plays an important role in determining not only the electronic structure but also the magnetic ordering.

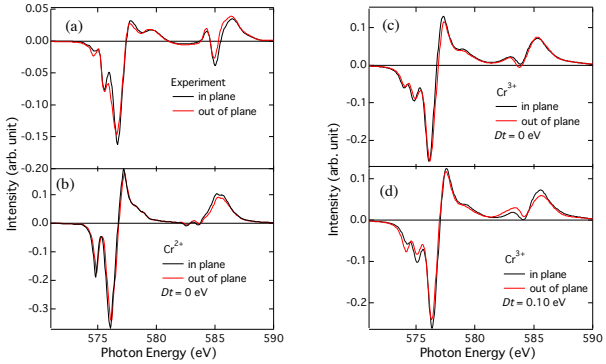

Fig. 3 XMCD spectra on CGT. (a) XMCD spectra of different direction of magnetic field. Cluster model calculation estimating the valence of Cr is 2+ (b) and 3+ (c), and 3+ with trigonal crystal field (d).

XMCD spectra of CGT are shown in Fig. 3. Differences in the spectra were observed between the different directions of magnetic field. In order to identify the origin of the spectral differences, we have conducted cluster-model calculation as shown in Figs. 3(b) – (d). The spectral line shape can be well reproduced by assuming Cr^{3+}, and not Cr^{2+}. Here, we include the trigonal crystal field (*Dt*) because $CrTe_6$ cluster is elongated along the c-axis, which makes the spectral line shape different with respect to the directions of magnetic field. This suggests that the distortion of $CrTe_6$ cluster is the origin of the difference.

For the future prospect, we would like to perform ARPES measurements for other vdW compounds such as $Cr_2Si_2Te_6$ in order to investigate the effect of the different ligand atoms, and also to perform a quantitative analysis of the XMCD spectra.

References

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