Direct Observation of the Three-dimensional Electronic Structure of RMnSi (R=La, Ce) with Noncentrosymmetic Antiferromagnetic Order

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Utilizing antiferromagnets for spintronics has recently attracted attentions. It is generally believed that it is difficult to control the physical properties of antiferromagnets by external fields due to the absence of macroscopic magnetization. However the magnetic multipole order hidden in the antiferromagnetism induces dramatic cross-correlational phenomena between electric and magnetic responses. Recently, a spintronic application using such antiferromagnetic order with hidden magnetic multipole has been proposed and has attracted considerable attention [1].

LaMnSi (CeMnSi) is an antiferromagnet with a Néel temperature of T_N ~ 293 K (~ 240 K). The crystal structure of RMnSi (R=La, Ce) belongs to the nonsymmorphic space group P4/nmm, as shown in Fig. 1(a) [2]. It contains a buckling layer composed of Mn and Si atoms as shown in Fig. 1(b). In the paramagnetic (PM) phase, the crystal does not have local inversion symmetry at each atomic site, leading to a sublattice connected by the spatial inversion symmetry operation [Fig. 1(c), Left]. Once the antiferromagnetic (AFM) order with q = 0 appears [Fig. 1(c), Right], the sublattice becomes inequivalent, and the global spatial inversion symmetry is spontaneously broken. This is prerequisite for the emergence of magnetic multipoles, which may lead to Rashba-type spin splitting and wavenumber-shifted asymmetric bands in the electronic structure [1], and may induce exotic external field responses [3]. In fact, magnetic piezoelectric effects have been observed in the antiferromagnets $BaMn_2As_2$ [4] and $EuMnBi_2$ [5], incorporating a buckling layer similar to that in Fig. 1(b), and the origin of the effects could be possibly ascribed to magnetic multipole order. In RMnSi, an anomaly in the electrical resistivity near T_N has been reported [2], suggesting a change in the electronic structure upon the AFM transition. It is noteworthy that RMnSi is different from ordinary antiferromagnets in that the magnetic moments are arranged in a q = 0 configuration without changing the size of the unit cell through the transition from the AFM phase with respect to the PM phase. This delivers an ideal stage to study

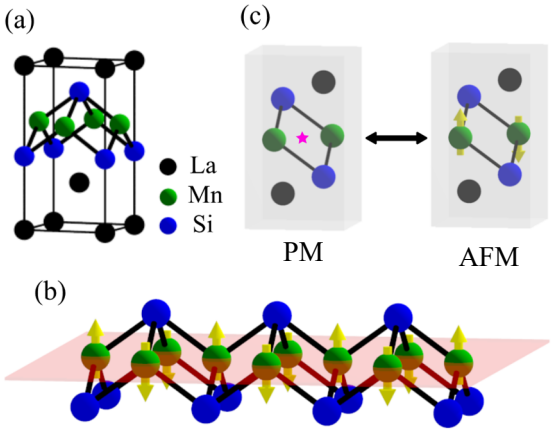

Fig. 1 (a) Crystal structure of LaMnSi. (b) MnSi layer in antiferromagnetic phase. (c) Structures of paramagnetic (Left) and antiferromagnetic states (Right).

the effect of magnetic ordering on the electronic structure. However, there are no experimental reports on the electronic structure of LaMnSi and CeMnSi, and it remains to be clarified whether the electronic structure reflects the symmetry due to the magnetic structure.

In this study, we performed angle-resolved photoemission spectroscopy (ARPES) using synchrotron radiation in the soft X-ray (SX) and vacuum ultraviolet (VUV) regions at SPring-8 BL25SU and HiSOR BL-1 / 9A, respectively, to clarify the effect of the symmetry of the AFM order on the electronic state in RMnSi (R = La, Ce). First, we observed the three-dimensional electronic structure including the band dispersion relation in the k_z direction using SX-ARPES, taking advantage of the tunable incident photon energy of synchrotron radiation. As shown in Fig. 2(a), the Fermi surface in the k_x-k_y plane including the Γ-X high symmetry line in the Brillouin zone was observed at T = 50 K below T_N. The band dispersion along the Γ-X line [cut in Fig. 2(a)] is shown in the left panel of Fig. 2(c). At the X point of the Brillouin zone boundary, a downward-convex parabolic band is clearly observed from the ARPES, and at the Γ point, the intersection of downward and upward dispersions is observed [Fig. 2(c), Right]. These features were also seen by the measurement using *p*-polarized VUV synchrotron radiation [Fig. 2(d)]. Similar dispersion was also observed for CeMnSi at 50 K (< T_N). Both of these experimental results were well reproduced by the first-principles calculation considering the antiferromagnetic order in LaMnSi than that for the PM phase [Fig. 2(c), Right]. These bands are mainly dominated by the Mn 3*d* orbital component, which implies that the Mn 3*d* electronic state responsible for antiferromagnetism has an itinerant character. Thus, the electronic states corresponding to the q = 0 AFM order in RMnSi (R = La, Ce) have been experimentally clarified for the first time.

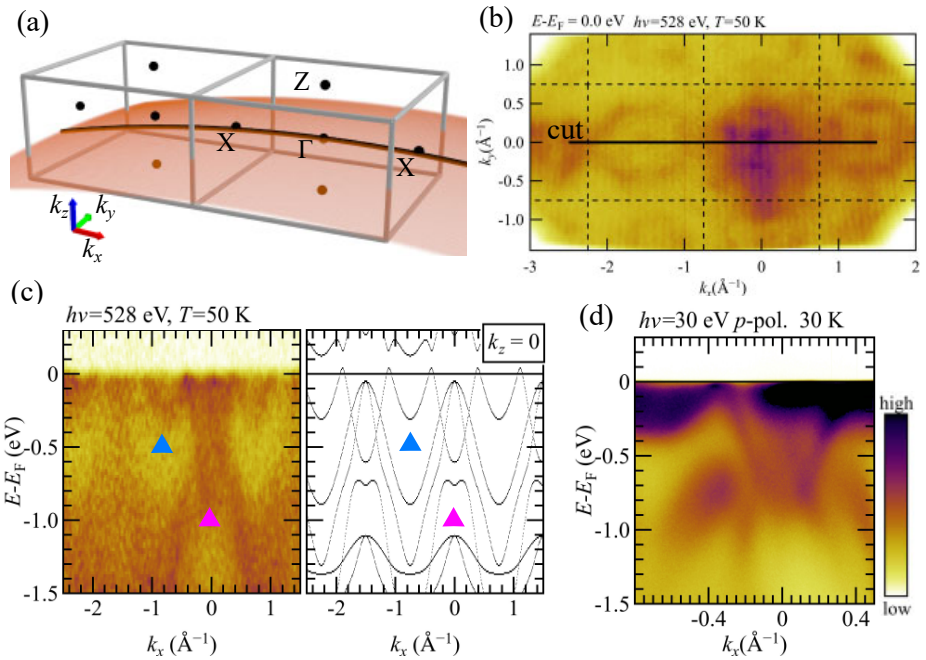

Fig. 2 (a) Brillouin zone of RMnSi. The red plane is the k_x-k_y cut plane at the incident photon energy $h\nu$=528 eV. (b) ARPES k_x-k_y inplane mapping of LaMnSi at 50 K (< T_N) at $h\nu$ = 528 eV at Fermi level. (c) Experimentally obtained energy dispersion ($h\nu$=528 eV) and first-principles calculation results in the Γ-X direction. (d) ARPES energy dispersion in the Γ-X direction for LaMnSi at $h\nu$ = 30 eV at 30 K (< T_N).

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