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μ- and SX-ARPES studies of compensated metal Cu_2Sb

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1 Introduction

Magnetoresistance (MR) is the magnetic-field induced changes of electrical resistance of a material, which has attracted great attention not only in understanding the underlying physical mechanisms but also for practical applications, such as spintronics devices, magnetic memory, and magnetic field sensors. The large MR has been considered to originate from nonsaturating behavior at high magnetic field, which is caused by either open Fermi surface or electron-hole compensation, in addition to high carrier mobility. Cu_2Sb is a layered compound composed of alternating stacks of Cu square net and CuSb block layer, and was reported to be a nonmagnetic metal exhibiting current-in-plane (CIP) MR of around 1000% at 4 K and 9 T more than 45 years ago. Such high MR was attributed to the open Fermi surface along the k_x direction. Although Cu_2Sb was suggested to be a compensated metal, the effect of electron-hole compensation in Cu_2Sb has been unclear since the fundamental electrical properties such as the carrier polarity, density, and mobility were not investigated. In addition, a quasi-two-dimensional (2D) Fermi surface confirmed by the de Hass–van Alphen effect and the band calculation indicated the presence of an open Fermi surface along the k_z direction, suggesting a possible large CPP-MR in Cu_2Sb. To uncover the electronic structure relevant to the MR effect, we have performed angle-resolved photoemission spectroscopy (ARPES) experiments on single crystals of Cu_2Sb.

2 Experiment

Cu_2Sb single crystals were grown by melt growth method with Cu_2Sb polycrystal which was synthesized by sintering mixed powders of Cu (99.99%) and Sb (99.99%). The crystal structure was evaluated by x-ray diffraction (XRD) with Cu Kα radiation (D8 DISCOVER, Bruker AXS).

VUV ARPES measurements were performed with a DA30 electron analyzer with micro-focused synchrotron light at BL-28 in the Photon Factory [2]. We used circularly polarized light of 60–80 eV. SX-ARPES measurements were performed at BL-2 (MUSASHI) in Photon Factory with 250–500 eV photons with horizontal linear polarization. The energy resolutions for VUV- and SX-ARPES measurements were set to be 10–30 and 150 meV, respectively. Samples were cleaved *in situ* along the a-b plane in an ultrahigh vacuum of 1×10^{-10} Torr, and kept at $T = 13$ K or 40 K during the measurements.

3 Results and Discussion

To determine three-dimensional (3D) bulk Fermi surface, ARPES measurements were performed at the normal-emission setup with varying photon energy ($h\nu$) in the SX region (300–375 eV). The ARPES-intensity mapping at E_F in the ΓMAZ plane shown in Fig. 1(a) reveals warped open Fermi surface whose periodicity well matches that of the bulk Brillouin zone. As shown in the corresponding momentum distribution curves (MDCs) at E_F in Fig. 1(b) plotted as a function of in-plane wave vector parallel to the ΓM cut ($k_{//}$), a few peaks originating from different Fermi surfaces are clearly resolved. A peak marked by a triangle apparently moves its **k** position upon $h\nu$ variation, demonstrating a finite k_z dispersion and thereby bulk origin of this Fermi surface. A careful look at Fig. 1(b) also reveals weak peaks (vertical dashed lines) whose **k** position is robust against $h\nu$ variation. These bands may be explained in terms of either the surface state or the k_z broadening effect occurring in the SX region. The quasi-2D nature of the pocket axially centered along the M-A line is also confirmed by the ARPES-intensity mapping at E_F plotted against in-plane wave vector (k_x and k_y) in Fig. 1(c),

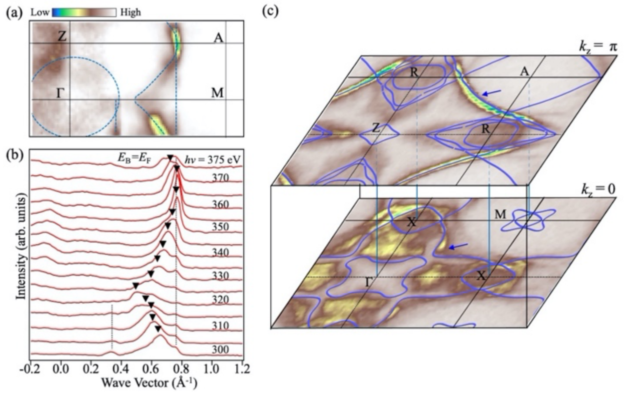

Fig. 1 (a) ARPES intensity at E_F plotted as a function of $k_{//}$ (parallel to the ΓM cut) and k_z measured by varying $h\nu$ with SX photons ($h\nu$ = 300–375 eV). (b) Corresponding MDCs at E_F. Triangles indicate the quasi-2D band, whereas dashed vertical lines show 2D-like bands. (c)ARPES-intensity mapping at E_F at k_z ~ 0 (bottom) and k_z ~ π (top). Calculated Fermi surfaces are also overlaid. Blue arrows indicate the quasi-2D pocket [1].

where one can recognize a large pocket at both k_z = 0 and π planes (blue allows) which are smoothly connected to each other upon variation of k_z [Fig. 1(a)]. Overall agreement of the ARPES-intensity distribution and the calculated band structure in Fig. 1(c) strongly suggests that the band calculation is a good starting point to describe the overall electronic structure of Cu_2Sb [1]. Although the existence of open Fermi surface was suggested from the previous studies, the present study unambiguously establishes that such Fermi surface indeed exists; this enables us to conclude that the Fermi-surface topology is correlated with the observed nonsaturating large CPP-MR.

Figures 2(a) and 2(b) show plots of ARPES-determined band dispersions measured along the high-symmetry cuts. The same plots but calculated band structure for k = 0 (solid curves) and π (dashed curves) are also superimposed. One can immediately recognize several sharp features in the ARPES intensity, supportive of the long quasiparticle lifetime, consistent with the high carrier mobility. Although the VUV data at $h\nu$ = 62 and 80 eV was expected to reflect information of k_z = 0 and π planes, respectively, based on the V_0 value (10.0 eV) estimated from the periodicity of the band dispersion in the SX data, the ARPES data at both $h\nu$'s suffer a strong k broadening effect. The calculation nicely reproduces the electron-like band axially centered along the M-A line which crosses E_F at the midway between the Z and A points or Γ and M cuts, being responsible for the aforementioned quasi-2D electron pocket. In addition, the holelike band crossing E_F around the Γ point forming the 3D hole pocket at Γ also shows a good matching between the experiment and calculation. These electron- and holelike bands show steep dispersion around E_F, contributing to the high carrier mobility. These results indicate that the existence of an open Fermi surface besides the electron-hole

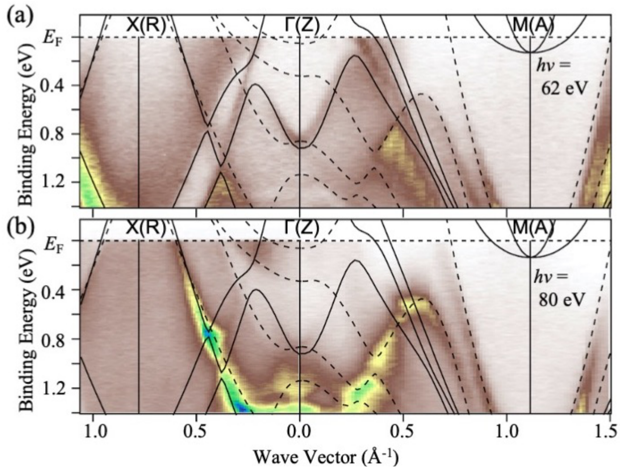

Fig. 2 (a) ARPES intensity plotted as a function of wave vector and binding energy measured along the XΓM cut (k_z = 0 plane) at $h\nu$ = 62 eV and the RZA cut (k_z = π plane) at $h\nu$ = 80 eV, respectively. Calculated band structures along the k_z = 0 and π is also overlaid with solid and dashed lines, respectively [1].

compensation is a key to realize large magnetoresistance in Cu_2Sb.

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