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Angle-resolved photoemission study of Sr_2RuO_4; An extended van-Hove singularity in non-cuprate superconductor

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We have performed high-resolution angle-resolved photoemission spectroscopy of a non-cuprate layered superconductor Sr_2RuO_4 to study the electronic structure near the Fermi level (E_F). The result shows an extended van Hove singularity (vHs) along the Ru-O bonding direction just like the cuprate, regardless of the character of the electronic states at E_F. This suggests that the extended vHs is a general electronic structure of a two dimensional correlated *d*-electron metal. We also have found that the observed Fermi surface topology (one electron-like Fermi surface sheet centered at Γ point and two hole-like ones centered at X point) is qualitatively different from the calculated Fermi surface topology (two electron-like Fermi sheets centered at Γ point and one hole-like one centered at X point) in the consequence of the existence of the vHs.

1. INTRODUCTION

The extended van-Hove singularity (vHs) derived from energy bands of the CuO_2 planes has been regarded as one of the characteristic electronic structure of high temperature superconductors (HTSC's), especially after the direct observation with high-resolution angle-resolved photoemission spectroscopy (HR-ARPES)[1-2]. Several models based on the vHs have been proposed to explain the high superconducting transition temperature and/or the T-linear dependence of the normal-state resistivity [3]. Sr_2RuO_4 is a non-cuprate layered Perovskite oxide superconductor (T_c ~ 1 K), having the same crystal structure as La_2CuO_4 with RuO_2 layers instead of CuO_2 in cuprate HTSC's [4]. The resistivity exhibits T-square (T^2) dependence below 25 K. This material would give us an unique opportunity to study the roll of CuO_2 plane.

In this paper, we report a high-resolution angle-resolved photoemission study of Sr_2RuO_4. We found an extended vHs near the Fermi level (E_F) in Sr_2RuO_4, which is quite similar to the vHs in cuprates, We discuss the correlation between the vHs and the superconducting and normal-state properties. We also mapped out the Fermi surface of Sr_2RuO_4.

2. EXPERIMENT

Single crystals were prepared by the floating zone method. X-ray diffraction was used to characterize the structure of the samples. Magnetic susceptibility measurements confirmed that superconductivity occurs below 0.9 K, as reported earlier [4]. HR-ARPES were carried out at the Synchrotron Radiation Center, Wisconsin, using a 4-meter normal incidence monochromator. The total energy and angular resolution were set at 20 meV (15 meV) and ±1°, respectively. The measurements were performed on several clean surfaces obtained by *in-situ* cleaving at 13 K with 21.2 eV photon energy. All spectra labeled in terms of the value of (k_x, k_y), where k_x and k_y are the wave vectors parallel to the Ru-O bonding direction (in units of $Å^{-1}$). A Pt foil in electrical contact with the sample was used as a Fermi level reference.

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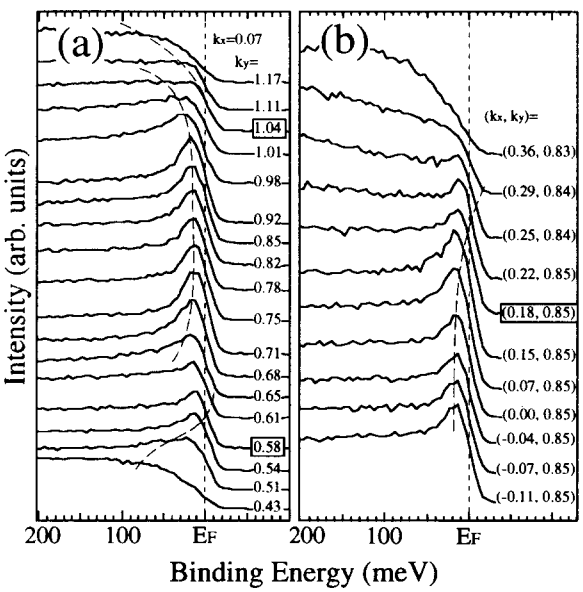
3. RESULTS AND DISCUSSION

Figures 1(a) and (b) show HR-ARPES spectra measured along ΓZ (Ru-O bonding direction) and XX in the Brillouin zone, respectively. Broken lines in the spectra are guide to the eyes. Measurement points in the Brillouin zone are shown with open and filled circles in Fig. 1(c), together with the calculated Fermi surface (dashed lines). In Fig. 1(a), we find that a band crosses E_F at k_y=0.58 and the same band enters from unoccupied states at k_y=1.04. But we do not observe further crossings between the two points. Instead, we find that a flat band is situated very close to E_F over a wide portion of the Brillouin zone centered around the mid point



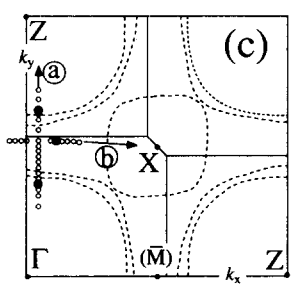


Fig. 1. HR-ARPES spectra near E_F along two directions (a) along ΓZ, and (b) along XX in the Brillouin zone. Measurement points in the Brillouin zone are shown with open and filled circles in Fig. 1(c).

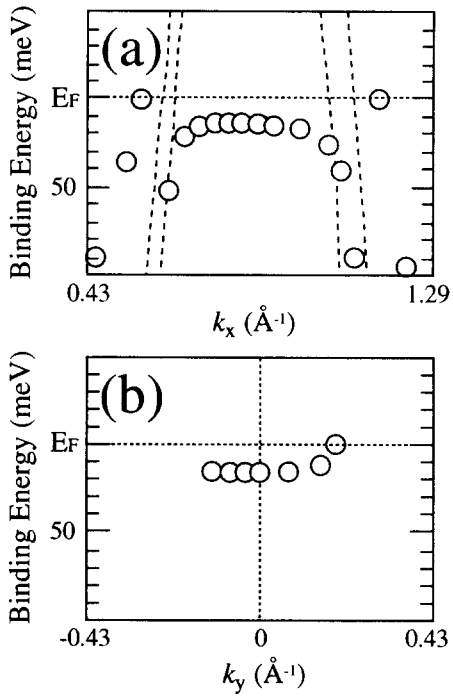


Fig. 2. Experimental band structure near E_F of Sr_2RuO_4 (open circles) determined by the present ARPES measurement. The band structure calculation [5] is shown by broken lines for comparison.

between the Γ and Z points (M point). To further characterize this flat band, we performed another set of HR-ARPES measurement along XX [Fig. 1(b)]. We find that a sharp peak just below E_F at [(k_x, k_y)=(0.00,0.85)] moves gradually toward E_F with increasing k_x and appears to enter into unoccupied states at [(k_x, k_y)=(0.18,0.85)].

The band dispersions obtained from the spectra in Figs. 1(a) and (b) are shown in Figs. 2(a) and (b), respectively. Dashed lines in Fig. 2(a) are the band calculation for comparison [5]. While two bands cross E_F in the calculation, only one band crosses it in the experiment as shown in Fig. 2(a). Another band near E_F shows a flat maximum over a substantial range of k values along ΓZ and also shows a flat minimum perpendicular to it, forming an extended saddle point. A similar extended vHs has been reported in several cuprate HTSC's [1-2]. For example, a HR-ARPES study of $YBa_2Cu_4O_8$ observed an extended vHs at 19 meV binding

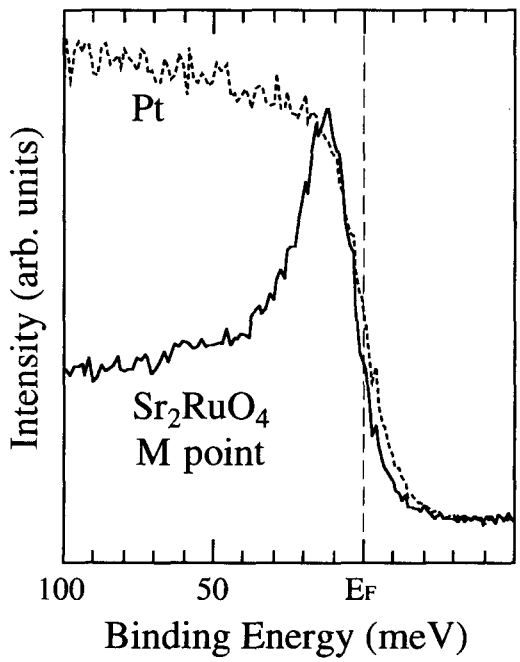


Fig. 3. Higher-resolution ARPES spectrum of Sr_2RuO_4 at the M point in the Brillouin zone measured at 13 K using 15 meV energy resolution, compared with that of a Pt foil measured under the same condition.

energy along the Cu-O bonding direction [2].

To accurately check the position of the extended vHs, we performed a higher-resolution measurement (15 meV total energy resolution) at the M point. As shown in Fig. 3, the peak is found at about 12 meV, and since its leading edge is sharper than the Fermi function as seen in the spectrum of Pt, it is clearly below E_F. The observation of the extended vHs with a smaller binding energy in a low T_c material raises a question as to the validity of the simple van-Hove scenario for high temperature superconductivity.

The van-Hove scenario also explains some anomalous normal-state properties such as the T-linear resistivity in terms of the critical energy of the vHs [3]. For example, King et al. have pointed out that $Nd_{2-x}Ce_xCuO_4$ shows T^2-dependent resistivity with its vHs located about 350 meV below E_F, while $Bi_2(Sr_{0.97}Pr_{0.03})_2CuO_6$ and Bi2212 show T-linear resistivity with vHs' being just below E_F [1]. However, in Sr_2RuO_4 this correlation does not hold. Therefore the simple explanation of anomalous normal-state properties in the van-Hove scenario has to be reexamined.

As a consequence of the presence of the extended vHs which is not predicted from the band calculation [5], the observed Fermi surface topology would be different from calculated one. In Fig. 4, we plot E_F-crossing points in the extended Brillouin zone determined by the HR-ARPES measurements (gray open, and filled circles). Dashed lines show the Fermi surface predicted by the band calculation [5]. The observed Fermi surface consists of one electron-like Fermi surface sheet centered at the Γ point and two hole-like ones centered at the X point. A remarkable difference between the experiment and the calculation is that one of the calculated electron-like sheets appears to transform into a hole-like sheet centered at the X point.

Such a drastic change in the Fermi surface topology could be lead from a change of the electron count, as discussed by Singh [6]. In Table 1, we listed the electron count of each and total Fermi surface sheets from our measurement (ARPES-1) and compare it with those from the dHvA result by

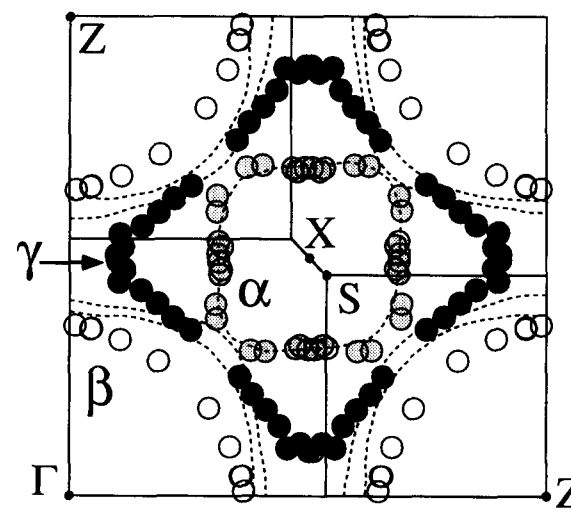


Fig. 4. Experimental Fermi surface of Sr_2RuO_4 obtained by the present ARPES measurement (α:gray, β:open and γ:filled circles). It consists of one electron-like Fermi surface sheet centered at the Γ point (β) and two hole-like ones centered at the X point (α and γ). The Fermi surface predicted by the band calculation [5] (broken lines) is also shown for comparison.

Table 1
Measured volumes of the electron count observed by three different measurements for Sr_2RuO_4:(a) present study, (b) A. P. Mackenzie et al.(dHvA) [7], and (c) D. H. Lu et al. (ARPES-2) [8]. The character of Fermi surface sheets obtained by the measurements is indicated in parenthesis (electron:e, hole:h).

	ARPES-1	dHvA	ARPES-2
α	1.75 ± 0.05 (h)	1.784 (h)	1.8 (h)
β	0.78 ± 0.08 (e)	0.914 (e)	0.7 (e)
γ	1.37 ± 0.09 (h)	1.334 (e)	1.5 (h)
Total	3.90 ± 0.22	4.032	4.0

Mackenzie et al.[7] and a recent ARPES result by Lu et al.[8] (ARPES-2). We found the total electron count from our measurement is 3.90±0.22, which is consistent with that expected for a Ru^{4+} ion and satisfies the Luttinger theorem. The electron count of each Fermi surface sheet shows a good agreement between the present ARPES study and the dHvA measurements, while the character (electron- or hole-like) of one Fermi surface sheet (γ Fermi sheet) is different between the two experiments. It is also noted that two independent ARPES studies show a good agreement with each other even for the character. The ARPES results seem consistent with the Hall coefficient measurement [9], which reported the sign change around 10 K from the negative to the positive value, indicative of the subtle balance of electron and hole counts as found by ARPES. The present result shows that the hole count (0.88±0.14) is close to the electron count (0.78±0.08). It is most likely that the electron correlations modify the band dispersion near E_F, and as a result give a different Fermi surface topology with an extended vHs just below E_F.

4. CONCLUSION

We have observed an extended vHs very close to E_F along the Ru-O bonding direction in analogy to the cuprate superconductors regardless of the difference of the character of band near E_F. This result suggests that an extended vHs may be a general feature of two-dimensional correlated metals. On the other hand, the large difference of T_c between $S_{r2}RuO_4$ and the cuprates, as well as qualitatively different transport properties, requests a reexamination of the van-Hove scenario and its relation to the normal and superconducting properties. As a consequence of the vHs, the experimental Fermi surface is different from that of the band calculation, although the total electron count is the same.

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REFERENCES

[1] D.M. King et al., Phys. Rev. Lett. 70 (1993) 3159; D.M. King et al., Phys. Rev. Lett. 73 (1994) 3298.
[2] K. Gofron et al., Phys. Rev. Lett. 73 (1994) 3302.
[3] D.M. Newns et al., Phyisica (Amsterdam) 186B (1993) 801.
[4] Y. Maeno et al., Nature 372 (1995) 532.
[5] T. Oguchi, Phys. Rev. B51 (1995) 1385.
[6] D.J. Singh, Phys. Rev B52 (1995) 1358.
[7] A.P. Mackenzie et al., Phys. Rev. Lett. 76 (1996) 3786.
[8] D.H. Lu et al., Phys. Rev. Lett. 76 (1996) 1845.
[9] N. Shirakawa et al., J. Phys. Soc. Jpn. 64 (1996) 1072.