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Fermi surface evolution of cobalt oxide Na_xCoO_2 studied by high-resolution angle-resolved photoemission spectroscopy

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Introduction

After the discovery of superconductivity in hydrated layered cobalt oxides ($Na_xCoO_2 \cdot yH_2O$) [1], intensive investigations have been performed to elucidate physical properties and their relation to the superconductivity. In particular, the antiferromagnetic (AF) transition in the highly doped region has attracted a special attention. In the AF phase, Co spins are ferromagnetically aligned within the in-plane and stacked antiferromaginetically along the *c*-axis. The inter-layer magnetic correlation is markedly strong as compared to other layered materials, suggesting an essentially three-dimensional (3D) nature of the magnetism, whereas the mechanism to trigger such anomalous AF state has not been well understood. It is thus indispensable to establish the electronic structure in highly Na-doped region to obtain insight into the microscopic origin of AF transition.

We have performed photon-energy-dependent angle-resolved photoemission spectroscopy (ARPES) on Na_xCoO_2 (x = 0.65 and 0.77). We have determined the 3D Fermi surface (FS) and band dispersion, and demonstrate that the FS topology shows a distinct transition as a function of Na doping.

Results and discussion

We show in Fig. 1 the photon-energy dependence of the ARPES spectra on the ΓALM plane of the Brillouin zone. Representative ARPES intensity is displayed in Fig. 2 [(a)-(e) for x = 0.77 and (g)-(k) for x = 0.65] [2]. At $h\nu$ = 55 and 65 eV, we observe a well-known highly dispersive holelike a_{1g} band which crosses E_F at $k_x \sim 0.6$ Å^{-1}. In sharp contrast, the band dispersion measured at $h\nu$ = 70 and 75 eV is unconventional: besides the holelike a_{1g} band at $k_x \sim 0.45$ Å^{-1}, an almost flat band appears in the close vicinity of E_F at -0.4 Å^{-1} ≤ k_x ≤ 0.4 Å^{-1}. Interestingly, this flat band again disappears at $h\nu$ = 85 eV. On the other hand, the $h\nu$ dependence of the ARPES spectra for x = 0.65 is surprisingly different. As shown in Figs. 1(g)-(k), we basically observe a single holelike band crossing E_F, and do not find any signature of the flat band. This indicates that the out-of-plane FS shape is considerably different between x = 0.65 and 0.77 samples. To see more clearly the FS shape, we plot in Fig. 1(f) and (l) the ARPES intensity at E_F as a function of k_x and k_z for x = 0.77 and 0.65, respectively. The FS of x = 0.65 slightly wiggles along the k_z direction, and the k_F position is closest (farthest) to k_x = 0 along the ΓM (AL) line. Such a wiggling is caused by a finite inter-plane electron hopping. At x = 0.77, we again find similar undulating behavior of FS, but its amplitude appears to be much pronounced. Moreover, we identify sizable ARPES intensity around the Γ point originating in the almost-flat band in Figs. 2(c) and (d), indicating the emergence of a new electron FS at Γ point. We concluded that appearance of this new FS is responsible to various anomalies in physical properties such as thermal conductivity and heat capacity.

References

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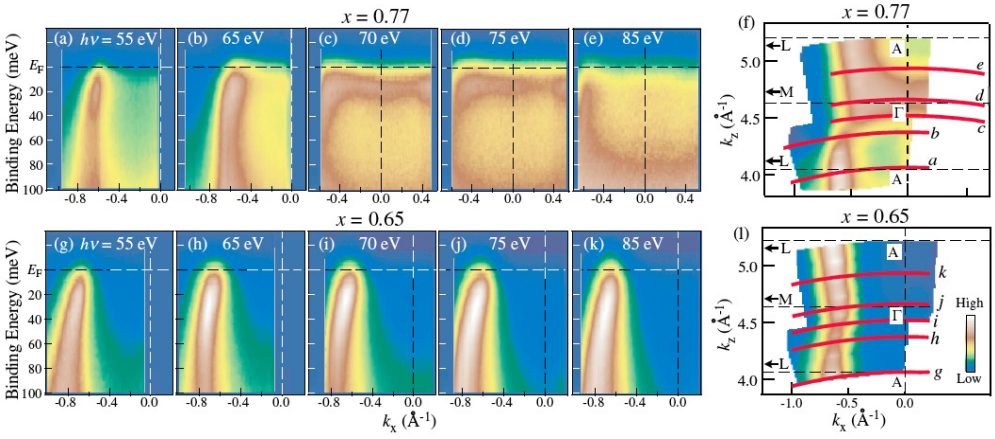

FIG.1: (a)-(e) hν dependence of ARPES intensity of $Na_{0.77}CoO_2$ as a function of k_x and binding energy along cuts *a-e* as indicated by red lines in (f). (f) ARPES intensity at E_F plotted as a function of k_x and k_z. Red lines show the momentum cuts where the spectra *a-e* were taken. (g)-(k) and (l): Same as (a)-(e) and (f) but for $Na_{0.65}CoO_2$.