Effects of Ru substitution on electron correlations and Fermi-surface dimensionality in Ba(Fe_{1-x}Ru_x)2As_2


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We report a systematic angle-resolved photoemission spectroscopy study on Ba(Fe_{1-x}Ru_x)2As_2 for a wide range of Ru concentrations (0.15 ≤ x ≤ 0.74). We observed a crossover from two dimensions to three dimensions for some of the holelike Fermi surfaces with Ru substitution and a large reduction in the mass renormalization close to optimal doping. These results suggest that isovalent Ru substitution has remarkable effects on the low-energy electron excitations, which are important for the evolution of superconductivity and antiferromagnetism in this system.

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I. INTRODUCTION

Superconductivity in the iron-based materials usually emerges from a magnetic state by several kinds of routes leading to very similar phase diagrams of magnetism and superconductivity. In BaFe2As2 and BaFe2CO3As2, the introduction of extra hole or electron carriers shifts the chemical potential so that the sizes of the hole and electron Fermi-surface (FS) pockets evolve oppositely, which eventually suppresses the nesting between the hole and electron FS pockets that play a role in the formation of a spin density wave (SDW) with exotic Dirac cone dispersion in the parent compound. While it is generally believed that external pressure also changes the FS topology by modifying the chemical bonds, the role of isovalent element substitution is still debated. Various scenarios, for example changes of the FS topology by chemical pressure, the reduction of electron correlations, magnetic dilution, and even the addition of extra hole carriers, have been suggested to explain the suppression of the SDW order in BaFe2As2 and BaFe2RuAs2 systems. Surprisingly, only little attention has been devoted to answer the reversed but somehow similar important question: How is superconductivity suppressed by increasing the substitution further than the optimal concentration?

Since single crystals can be grown for the entire phase range wider than in previous studies. Our data indicate that the impact of the degradation of the nesting between hole and electron FS pockets cannot be neglected. However, the most significant changes in the electronic band structure are observed at optimal concentration and at higher Ru content. We reveal a clear crossover from two dimensions to three dimensions for some of the holelike FSs with Ru substitution accompanied by a reduction of mass renormalization, suggesting that the isovalent Ru substitution has significant effects on the low-energy electron excitations. The implications of the changes in the FS topology and the near-\EF band dispersions on the evolution of the SDW order and superconductivity are discussed.

II. EXPERIMENT

High-quality single crystals of Ba(Fe_{1-x}Ru_x)2As_2 were grown by the flux method. The Ru concentration of the samples was determined by energy-dispersive spectroscopy x-ray and x-ray diffraction analyses with an accuracy of ~0.01. ARPS experiments were performed at beamlines PGM and Apple-PGM of the Synchrotron Radiation Center (Wisconsin), beamline 12.0.1 of the Advanced Light Source (California), and beamline SIS of the Swiss Light Source (Switzerland) with Scienta analyzers. The energy and angular resolutions were set at 20 meV and 0.2°, respectively. The samples were cleaved \textit{in situ} and measured at 20 K in a vacuum better than 4 × 10^{-11} Torr.
FIG. 1. (Color online) (a) Temperature dependence of the in-plane resistivity of Ba(Fe_{1-x}Ru_{x})_2As_2 single crystals for x = 0.15, 0.21, 0.33, 0.39, 0.44, 0.51, and 0.74 respectively (named Ru15, Ru21, Ru33, Ru39, Ru44, Ru51, and Ru74). (b) Phase diagram of Ba(Fe_{1-x}Ru_{x})_2As_2 based on the resistivity data. (c) Core levels recorded with a photon energy of 195 eV. (d) Doping dependence of the Fe 3p (E_F ∼ 53 eV) peak weight. (e)–(g) Intensity plot along the high symmetry lines Z-A-R-Z for Ru15, Ru39, and Ru74. Nonrenormalized LDA bands are also plotted for comparison.

III. RESULTS AND DISCUSSION

The normalized resistivity data plotted in Fig. 1(a) suggest that the SDW order identified as a resistive anomaly (upturn) is gradually shifted to low temperature with Ru substitution and disappears for x ≥ 0.30. Superconductivity emerges for x > 0.15, reaches its maximum transition temperature at x ∼ 0.30, and is strongly suppressed for x > 0.39. The SDW temperature T_{SDW} and critical temperature T_c extracted from the resistivity data are plotted in the T-x phase diagram, Fig. 1(b). Figure 1(c) shows the shallow core levels and the valence band spectra for different Ru concentrations. The peak position of the Fe 3p orbital at the binding energy (E_F) of ∼53 eV is not shifted with Ru substitution, confirming that the valence state of the Fe ions is not changed. Furthermore, the intensity of the Fe 3p peak extracted by integrating the peak area decreases linearly as a function of the Ru concentration and can be extrapolated to zero for BaRu2As2, as shown in Fig. 1(d). In contrast, the Ru 4p peak at E_F ∼ 48 eV is enhanced with increasing the Ru concentration, though it is too weak to extract accurately its intensity value.

In iron-based superconductors, most of the dispersive bands originating from the Fe 3d orbitals are located within 2 eV below E_F. To illustrate the effects of Ru substitution on the dispersive bands, we plot in Figs. 1(e)–1(g) the photoemission intensity along the high symmetry lines Z-A-R-Z for x = 0.15, 0.39, and 0.74, respectively. For comparison, we also plot the band structure within the local density approximation (LDA) for x = 0, 0.4, and 1 in Figs. 1(e)–1(g), respectively. While the overall bandwidth increases slightly for higher Ru concentrations as expected from band calculations, it is interesting to notice that the calculated bands do not need to be renormalized to fit the dispersive bands in the high E_F range. In contrast, the near-E_F bands are strongly renormalized, especially for x ≤ 0.39, suggesting that the many interactions mainly affect the low-energy electron excitations.

In order to clarify the effects of Ru substitution on the low-energy excitations, we focus on the band dispersions near E_F. Figure 2(a) shows a photoemission intensity plot at the Γ point (k_z = 0) for Ru15, taken in the s-pol geometry. (b) Corresponding energy distribution curves. (c)–(e) Curvature plot of the momentum distribution curves (MDCs) along Γ-M (k_z = 0) in the s-pol geometry. (f)–(h) Same as (c)–(e), but along the Z-A (k_z = π) symmetry line. (i) Same as (d), but in p-pol geometry. (j) Same as (g), but in p-pol geometry.

FIG. 2. (Color online) (a) ARPES intensity plot along Γ-M (k_z = 0) for Ru15, taken in the s-pol geometry. (b) Corresponding energy distribution curves. (c)–(e) Curvature plot of the momentum distribution curves (MDCs) along Γ-M (k_z = 0) in the s-pol geometry. (f)–(h) Same as (c)–(e), but along the Z-A (k_z = π) symmetry line. (i) Same as (d), but in p-pol geometry. (j) Same as (g), but in p-pol geometry.
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FIG. 3. (Color online) (a)–(c) Hole FS maps in $k_z$-$k_y$ plane in $s$-pol geometry. (d) Same as (a), but in the $p$-pol geometry. (e) Electron FS maps in $k_z$-$k_y$ plane for Ru21 in the $s$-pol geometry. (f) Same as (e), but obtained in the $p$-pol geometry. (g)–(i) Fermi momenta (symbols) of the hole bands. (j) Fermi momenta of the electron bands (symbols).

respectively. One can see two FSs clearly in the $s$-pol geometry. One of them (hereafter called $\alpha$) keeps a quasi-2D behavior for all Ru concentrations whereas the other one (called $\alpha'$) exhibits clearly a crossover from 2D to 3D with Ru substitution. Figure 3(d) shows the $p$-pol geometry data for Ru15, in which one can see a third FS (called $\alpha'$). As shown in Figs. 3(e) and 3(f), two quasi-2D electronlike FSs are distinguished at the M point in both the $s$-pol and $p$-pol geometries, with no obvious change upon Ru substitution.

The $k_F$ of the three hole bands extracted from the MDCs are plotted in Figs. 3(g)–3(i), while the $k_F$ for the two electron bands are given in Fig. 3(j). The results, as well as the total number of carriers estimated from the Luttinger theorem, are displayed as a function of the Ru concentration in Fig. 4(a). The hole carriers are well compensated by the electron ones at all Ru concentrations, demonstrating that the Ru substitution is isovalent, in agreement with the core-level measurements.

Although other factors such as magnetic dilution should be taken in account, our experimental results thus suggest that the degradation of the nesting conditions may play a role in the suppression of the SDW ordering with Ru increasing.

To further illustrate the effects of the Ru substitution on the low-energy excitations, we compare the extracted dispersions of the holelike and the electronlike bands at $k_z = \pi$ for $x = 0.15$ and 0.74 in Figs. 4(b) and 4(c), respectively. The dispersions of all the bands for $x = 0.74$ are much steeper than those for $x = 0.15$. Figure 4(d) displays the extracted Fermi velocity ($v_F$) values at $k_z = \pi$ as a function of Ru concentration. While the $v_F$ values in the $k_z = \pi$ plane are not obviously changed in the underdoped range, they increase rapidly in the overdoped range. This effect is captured by our LDA band structure calculations, for which the $v_F$ values corresponding to the $\gamma$ band in the $k_z = \pi$ plane are shown in Fig. 4(d). Interestingly, the large increase in the $v_F$ values in the overdoped regime is accompanied by an important decrease in the mass renormalization. We note that these sudden changes in $v_F$ and in the band renormalization occur near optimal doping. From the phase diagram given in Fig. 4(d), one can see that the optimal doping for superconductivity is also close to the phase boundary of the SDW state. It is interesting to point out that similar trends in mass enhancement toward the boundary of the magnetic order phase have also been revealed in another isovalent substituted system, BaFe$_2$(As$_{1-x}$P$_x$)$_2$, and in the heavy-fermion system CeRhIn$_5$ by de Haas-van Alphen experiments. This gives us a hint that the many-body interactions, which give rise to the enhancement of the effective mass of the quasiparticles in the low doping regime, could be associated with the magnetic quantum critical point (QCP) and could play an important role in the unconventional superconductivity of these systems.

Whether a localized or an itinerant picture is most appropriate to describe superconductivity in the Fe-based superconductors is still a matter of controversy. Let’s first assume that an itinerant picture prevails. In Figs. 4(e)–4(f), we plot the 3D hole FSs for underdoped and optimally doped Ba(Fe$_{1-x}$Ru$_x$)$_2$As$_2$. Our results show that the holelike $\beta$ FS remains quasi-2D and is still quasinested with the electronlike FSs far beyond the superconducting dome. In contrast, the $\alpha$ and $\alpha'$ FSs become very 3D with Ru substitution. The tops of
the $\alpha$ and $\alpha'$ bands are degenerated at $\Gamma$ [see Figs. 2(c) and 2(i)], suggesting that they mainly originate from the $d_{z^2}$ orbitals, in agreement with a recent ARPES study on the (Ba,K)Fe$_2$As$_2$ system.\textsuperscript{21} The enhancement of the 3D character is attributed to $\alpha$ the electronic structure of Ru 4$d$ due to the enhanced spacial extension of the Ru 4$d$ orbitals and a decrease of the As height.\textsuperscript{6,12,13} On the other hand, the quasi-2D $\beta$ FS is attributed to the $d_{xy}$ orbital, whose spatial extension is limited to the $a$-$b$ plane. Our results indicate clearly that if quasinesting between the hole and electron FS pockets is the determinant factor for superconductivity in these materials, then the $d_{z^2}$ orbitals are mainly responsible. Within a local picture, the main interpretation for the suppression of superconductivity at high Ru content can be explained in terms of both the reduction of the electronic correlations reported in this paper and the important modifications of the local antiferromagnetic exchange constants due to the increased spacial extension of the Ru 4$d$ orbitals and their increased hybridization with the As 3$p$ orbitals due to the decrease of the As height.

IV. SUMMARY

In summary, our study of Ba(Fe$_{1-x}$Ru$_x$)$_2$As$_2$ over a wide range of doping indicates that the degradation of the nesting conditions between the hole and electron FS pockets cannot be neglected in understanding the suppression of the SDW in this system. However, the main changes in the electronic band structure occur for Ru concentrations exceeding optimal substitution. In that regime, some hole bands become much more 3D, an effect accompanied by an increase of the Fermi velocities and a strong suppression of the electronic correlations. Our study suggests that one or many of these strong effects may play a role in the suppression of superconductivity in the high-substitution regime of Ba(Fe$_{1-x}$Ru$_x$)$_2$As$_2$.

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