Is $\text{BaCr}_2\text{As}_2$ symmetrical to $\text{BaFe}_2\text{As}_2$ with respect to half 3$d$ shell filling?

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We have performed an angle-resolved photoemission spectroscopy study of $\text{BaCr}_2\text{As}_2$, which has the same crystal structure as $\text{BaFe}_2\text{As}_2$, a parent compound of Fe-based superconductors. We determine the Fermi surface of this material and its band dispersion over 5 eV of binding energy. Very moderate band renormalization (1.35) is observed for only two bands. We attribute this small renormalization to enhanced direct exchange as compared to Fe in $\text{BaFe}_2\text{As}_2$, and to a larger contribution of the $e_g$ orbitals in the composition of the bands forming the Fermi surface, leading to an effective valence count that is reduced by Fe $d$ - As $p$ hybridization.

I. INTRODUCTION

The electronic correlations are widely believed to play a major role for unconventional superconductivity in the Fe-based superconductors [1]. It was established theoretically that the strength of the electronic correlations in these materials is tuned by the filling of the 3$d$ shell in presence of a large Hund’s coupling [2-7]. Accordingly, angle-resolved photoemission spectroscopy (ARPES) studies report that the band renormalization factor $1/Z$ decreases monotonically upon filling the 3$d$ shell: $1/Z = 3$ in $\text{BaFe}_2\text{As}_2$ ($d^8$) [8] (consistent with density functional theory (DFT) + dynamic mean-field theory (DMFT) calculations [9]), $1/Z = 1.4$ in $\text{BaCo}_2\text{As}_2$ ($d^7$) [10,11], $1/Z = 1.1$ in $\text{SrNi}_2\text{As}_2$ ($d^8$) [12], and $1/Z = 1$ in $\text{BaCr}_2\text{As}_2$ ($d^{10}$) [13]. Within this framework, the electronic correlations should evolve similarly as a function of band filling with respect to the half 3$d$ shell case ($d^8$). Therefore, the $d^4$ case of $\text{Cr}^{2+}$, the symmetric counterpart of the $d^6$ filling of Fe$^{2+}$, raises the possibility of unconventional superconductivity for compositions in proximity of $\text{BaCr}_2\text{As}_2$. As with $\text{BaFe}_2\text{As}_2$, $\text{BaCr}_2\text{As}_2$ is an antiferromagnetic metal, and a sizable renormalization factor of 2 was derived from specific heat measurements [14]. A recent DMFT study also suggests mass enhancement by a factor of 2 [15], thus reinforcing the view that $\text{BaCr}_2\text{As}_2$ can be regarded as the symmetrical analog of $\text{BaFe}_2\text{As}_2$. A possible pairing instability is even proposed upon negative pressure or electron doping [15]. However, there is to date no direct experimental characterization of the electronic band dispersion of $\text{BaCr}_2\text{As}_2$ [16], which is essential to answer the question: Is $\text{BaCr}_2\text{As}_2$ symmetrical to $\text{BaFe}_2\text{As}_2$ with respect to half 3$d$ shell filling?

Here we present an ARPES study of $\text{BaCr}_2\text{As}_2$. Despite evidence for a surface state, the Fermi surface (FS) is very similar to the one expected from local density approximation (LDA) calculations. We find that while most bands are not renormalized, two bands near the Fermi energy ($E_F$) are renormalized by a factor 1.35, which is much smaller than reported theoretically. We show that unlike in $\text{BaFe}_2\text{As}_2$, the spectral weight at $E_F$ in $\text{BaCr}_2\text{As}_2$ is significantly contaminated by $e_g$ orbitals and that direct exchange between Fe neighboring atoms play a more significant role. From our experimental results and analysis, we conclude that although there are obvious similarities between $\text{BaCr}_2\text{As}_2$ and $\text{BaFe}_2\text{As}_2$, the analogy between these two compounds cannot be pushed too far.

II. METHODS

High-quality single crystals of $\text{BaCr}_2\text{As}_2$ were grown by the self-flux method [14]. ARPES measurements were performed using photon energies ($h\nu$) of 56 eV and 73 eV at Beamline I05 of Diamond Light Source equipped with a VG-Scienta R4000 analyzer. The energy and angular resolutions were set at 12 - 15 meV and 0.2°, respectively. Additional measurements in the 22 - 80 eV $h\nu$ range have been recorded at the PGM beamline of the Synchrotron Radiation Center equipped with a VG-Scienta R4000 analyzer, with the energy and angular resolutions set at 20 - 30 meV and 0.2°, respectively. All samples were cleaved in situ and measured at 20 K in a vacuum better

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We have performed DFT calculations within the LDA and in the G-type antiferromagnetic order, as implemented in the Wien2k code [17]. The lattice parameters were taken from experiment (a = 3.963 Å, c = 13.600 Å) [18] and we chose \( z_{As} = 0.3572 \), similar to what was found in the calculations of Singh et al. [14].

than 5 \times 10^{-11} \text{Torr}. Throughout the paper, we label the momentum values with respect to the 1 Cr/unit-cell Brillouin zone (BZ), and use \( c' = c/2 \) as the distance between two Cr planes.

III. RESULTS AND DISCUSSION

In Figs. 1(a) and 1(b), we display our calculations of the FS of BaCr\(_2\)As\(_2\) in the antiferromagnetic state for \( k_z = 0 \) and \( k_z = \pi \), respectively. Our results are consistent with previous calculations [14, 15] and show that the FS differs substantially from that of the Fe-based superconductors. The calculations indicate the existence of 3 hole FS pockets centered at the \( \Gamma \) point \((0,0,0)\), and 2 hole FS pockets centered at the \( Z \) point \((\pi,\pi,0) = (0,0,\pi)\). Here we call \( \alpha \) the inner FS pocket, which is 3D and does not appear around the \( Z \) point. The \( \beta \) band, which forms the \( \beta \) FS in the \( k_z = 0 \) plane, disperses only slightly along \( k_z \) and gives the \( \beta' \) FS pocket around the \( Z \) point. In contrast, the \( \gamma \) band varies more strongly along \( k_z \) to give a \( \gamma \) FS pocket in the \( k_z = 0 \) plane and a \( \gamma' \) FS pocket in the \( k_z = \pi \) plane that have different shapes and sizes.

The orbital projections of the calculated band dispersions are shown in Fig. 2 with the weight of the majority spin (Cr1) and minority spin (Cr2) plotted separately. Our calculations indicate that both the \( \alpha \) and \( \beta \) FSs derive mainly from Cr1 \( d_{xz} + d_{yz} \), but that their orbital composition is not pure and includes other Cr1 and Cr2 \( d \) orbitals as well. In contrast, the \( \gamma \) band derives almost only from Cr1 \( d_{xy} \). Although most of the As \( p \) states
locate below $E - E_F = -1.5$ eV, our calculations also predict some weight for As $p_z$ at $E_F$.

The experimental sections of FS displayed in Figs. 1(c) and 1(e) for $k_z \approx 0$, and in Figs. 1(d) and 1(f) for $k_z \approx \pi$, are very similar to the calculations. This is notably true for the $\gamma$ and $\gamma'$ FS pockets. Small discrepancies need to be reported though. For example, while the calculations predict a slightly octagonalish shape for the section of the $\beta$ FS around $\Gamma$, what is observed experimentally is more squarish. The $\alpha$ FS in Figs. 1(c) and 1(e) is also circular, in contrast to the rather squarish calculated one. More importantly, a replica of that FS pocket is detected at the $Z$ point, suggesting a band folding or a surface state.

Our analysis shows that the $\alpha$, $\beta$, and $\gamma$ sections of FS cover respectively 7.4 %, 34.5% and 63.3% of the 2Cr/unit-cell BZ (dashed red squares in Fig. 1). At $k_z \approx \pi$, the $\beta'$ and $\gamma'$ sections of FS cover 50.2 % and 35.1 % of the 2Cr/unit-cell BZ. After averaging the total areas at $k_z \approx 0$ and $k_z \approx \pi$, we find 95.3 % of the 2Cr/unit-cell BZ. Considering spin degeneracy, this coincides to slightly less than the expected $d^4$ filling of Cr$^{2+}$. Our approximation is justified by the $k_y - k_z$ mapping showed in Fig. 3(m), obtained by scanning $h\nu$ from 22 to 80 eV. The conversion from $h\nu$ to $k_z$ is done using the three-step model with the sudden approximation and the free-electron final state, and we used an inner potential of 13 eV [19]. The $\alpha$ FS, small compared to the others, is dispensionless, but has stronger intensity around the $\Gamma$ point, giving further support for the existence of a surface state. While the $\beta$ band is nearly dispensionless along $k_z$, the $\gamma$ band shows sizable dispersion along $k_z$.

In Figs. 3(a) and 3(b) we show the electronic band dispersion observed experimentally from ARPES intensity cuts along $\Gamma$-M ($k_z = 0$) recorded using 73 eV photons ($k_z \approx 0$) in the $s$ and $p$ polarization configurations, respectively. In a similar fashion, we display in Figs. 3(g) and 3(h) the ARPES intensity cuts recorded with 56 eV photons ($k_z \approx \pi$), corresponding to the Z-A high-symmetry line. The LDA calculations are superimposed for comparison. The curvature intensity plots [21] and the energy distribution curves (EDCs) corresponding to the ARPES cuts are displayed on their right side. The agreement between experiments and calculations is rather good over 4.5 eV and most bands would need only small energy shifts to match the ARPES intensity plot. While the $s$ polarization configuration favors the observation of states with odd symmetry or with $z$-oriented orbitals such as $d_{x^2}$ or $p_z$, the $p$ polarization configuration favors even symmetry states. Accordingly, the $\gamma$ ($d_{xy}$) band is more intense under $s$ polarization. The experimental results also indicate that the $\beta$ and $\alpha$ bands have dominant odd and even symmetries, respectively.

The most obvious difference between the calculations and the experimental results is the presence of the band labeled $\zeta_2$ in Fig. 3 which is not predicted. As with the $\zeta_1$ band associated to the Cr1 $d_{z^2}$ orbital, it appears clearly only under $s$ polarization. Interestingly, the dispersions of the $\zeta_1$ and $\zeta_2$ bands are identical within uncertainties, and the location of the bottom of the $\zeta_2$ band is nearly the same under both 56 eV and 73 eV photon excitations. Consequently, we conjecture that the $\zeta_2$ band is a surface state with $d_{z^2}$ orbital character.

As mentioned above, the unrenormalized LDA calculations are in good agreement with the experimental data.
orbital fillings. In fact, another – possibly more impor-
tant – factor is the orbital-resolved kinetic energy that
is larger for orbitals having a low density-of-states at the
Fermi level. As discussed in Ref. [3] for FeSe, the or-
bitals that exhibit a single-particle pseudogap are less
correlated. This can be rationalized by the presence of
more possibilities for fluctuations around the Fermi level
in the other three orbitals. A similar mechanism could
be at work in BaCr$_2$As$_2$.

We now try to understand why the 122 structure leads
to superconductivity with a high superconducting transi-
tion temperature ($T_c$) for Fe-based compounds, whereas
no superconductivity is reported for their Cr-based coun-
terparts. The strength of the electronic correlations,
which we show in this work to be very different in these
two systems, may play an important role. Possible su-
perconductivity was predicted for 3$d^n$ fillings somewhere
between $n = 4$ and $n = 5$ [15, 22], where the strength of
the electronic correlations should be comparable to that
in Fe-based superconductors. Yet, additional issues need
to be addressed. For instance, the magnetic ordering is
different in the two systems. BaCr$_2$As$_2$ shows a $G$-type
ordering, and a Néel temperature of 580 K has been re-
ported [25], with Cr spins pointing along the $c$-axis in
EuCr$_2$As$_2$ [26]. In contrast, BaFe$_2$As$_2$ orders antiferro-
magnetically around 140 K with spins aligned inside the
Fe plane in a bi-collinear structure [27]. The antifer-
romagnetic wave vector in the Fe-based superconductors
connects hole and electron pockets FS pockets, giving rise
to the quasi-nesting scenario suggesting that low-energy
spin fluctuations may contribute to the superconducting
pairing [25, 30]. Although the quasi-nesting scenario is
now seriously challenged [19], in particular due to the
existence of high-$T_c$ superconductivity without hole FS
pocket in $A_x$Fe$_{2-y}$Se$_2$ [31, 32] and FeSe monolayers [35-
39], the $\Gamma$-$M$ wave vectors connects the top of the
holelike bands and the bottom of the electron bands within an
energy range smaller than a few hundreds of meV in all Fe-
based superconductors, suggesting that high-energy spin
fluctuations (local moments) may play a role. There is no
such electron-hole connection with the antiferromagnetic
wave vector in BaCr$_2$As$_2$.

Even the $\alpha$ band, near $E_F$, is well reproduced by the
renormalized LDA calculations. However, the situation
is not as good for the $\beta$ and $\gamma$ bands. As we show in Fig.
4 by zooming on the band dispersion near $E_F$, these two
bands fit the calculations better after renormalizing the
LDA bands by a factor of $1.35$. This factor is far from
that observed in the Fe-based superconductors and more
consistent with the renormalization found in BaCo$_2$As$_2$
[10, 11], which corresponds to a $d^7$ filling. From that per-
spective, the case of BaCr$_2$As$_2$ is thus not symmetrical
to BaFe$_2$As$_2$. A similar asymmetry was found theoreti-
cally [22] in the 1111 phase (corresponding to LaFeAsO
and LaCrAsO). There, it was attributed to the orbital-
dependence of the electronic filling [23, 24]. Indeed, the
t$_{2g}$ orbitals ($d_{xz}/d_{yz}$ and $d_{xy}$) are closer to half-filling
for the $d^6$ configuration (Fe) than for a $d^4$ filling (Cr),
whereas the less correlated e$_{2g}$ orbitals ($d_{z^2}$ and
$d_{x^2-y^2}$) are the ones closest to half-filling for the $d^4$
configuration [22]. In that picture, recovering a similar electronic mass
enhancement as in LaFeAsO would require a $d^n$ filling
of around $n = 4.6$ to 4.8. On the other hand, the sit-
uation might not be only determined by the respective
orbital fillings. In fact, another – possibly more impor-

FIG. 4. (Color online). (a) Near-$E_F$ zoom of the curva-
ture intensity plot obtained with s-polarization at $k_z \approx 0$.
Dashed lines in red and in purple represent unrenormalized
LDA bands and LDA bands renormalized by a factor 1.35, re-
spectively. (b) Same as (a) but recorded with p polariza-
tion at $k_z \approx 0$. (c) Same as (a) but recorded with s polarization
at $k_z \approx \pi$. (d) Same as (a) but recorded with p polarization
at $k_z \approx \pi$. (e) Same as (a) but recorded with p polarization
at $k_z \approx \pi$.
With that respect, BaFe$_2$As$_2$ and BaCr$_2$As$_2$ are very different. Finally, one might speculate that the enhanced $d$-$p$ hybridization leads to an effectively reduced $d$-electron count in BaCr$_2$As$_2$, which would then be consistent with a correlation strength comparable to the $d^7$ compound BaCo$_2$As$_2$. A similar mechanism was indeed discussed in Ref. [47] where it was highlighted that the correlation strength depends on the effectively available charge rather than on the nominal $d$-electron count. Finally, one might also speculate that the increased hybridization also influences the effective crystal field splittings (through ligand field effects) thus modifying more profoundly the scenario of the Hund’s metal phase appearing around the $d^6$ filling.

IV. SUMMARY

In summary, we performed an ARPES study of BaCr$_2$As$_2$ that indicates good consistency with LDA calculations in the antiferromagnetic state. A renormalization factor of 1.35 is observed for only two bands, indicating that BaCr$_2$As$_2$ is much less correlated than BaFe$_2$As$_2$. Our analysis suggests a stronger weight of the $e_g$ orbitals at $E_F$, which may be responsible for the observed asymmetry with respect to half 3$d$ shell filling.

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