

## Evidence of Strong Correlations and Coherence-Incoherence Crossover in the Iron Pnictide Superconductor $\text{KFe}_2\text{As}_2$

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Using resistivity, heat-capacity, thermal-expansion, and susceptibility measurements we study the normal-state behavior of  $\text{KFe}_2\text{As}_2$ . Both the Sommerfeld coefficient ( $\gamma \approx 103 \text{ mJ mol}^{-1} \text{ K}^{-2}$ ) and the Pauli susceptibility ( $\chi \approx 4 \times 10^{-4}$ ) are strongly enhanced, which confirm the existence of heavy quasiparticles inferred from previous de Haas–van Alphen and angle-resolved photoemission spectroscopy experiments. We discuss this large enhancement using a Gutzwiller slave-boson mean-field calculation, which shows the proximity of  $\text{KFe}_2\text{As}_2$  to an orbital-selective Mott transition. The temperature dependence of the magnetic susceptibility and the thermal expansion provide strong experimental evidence for the existence of a coherence-incoherence crossover, similar to what is found in heavy fermion and ruthenate compounds, due to Hund’s coupling between orbitals.

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Soon after the discovery of high-temperature superconductivity in iron pnictide compounds [1,2], their unique electronic structure, displaying electron and hole sheets, was revealed. In the  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  series, a broad superconducting dome emerges in the vicinity of an antiferromagnetic spin density wave (SDW) instability ( $x \approx 0.3$ ) and is maximal at  $x \approx 0.4$  with  $T_c \approx 38 \text{ K}$ . At this optimal concentration, the superconducting order parameter is fully gapped with either  $s^{++}$  or  $s^{+-}$  symmetry [3,4]. In the latter case, it is believed that pairing is due to repulsive interband interactions enhanced by the magnetic fluctuations which develop around the nesting vector that connects the two different sheets [3]. However, superconductivity is not confined to this hypothetical quantum critical region and persists to  $x = 1$  (with a strongly depressed  $T_c \approx 3 \text{ K}$ ) [2], where only hole pockets are present [5–9]. Moreover, the Sommerfeld coefficient for  $\text{KFe}_2\text{As}_2$ ,  $\gamma \approx 100 \text{ mJ mol}^{-1} \text{ K}^{-2}$ , is paradoxically about twice larger than observed at the optimal concentration [10,11] and recent laser angle-resolved photoemission spectroscopy (ARPES) measurements reveal that some of the energy gaps have nodes [12]. Clearly, more experimental investigations are necessary to elucidate the situation in the overdoped region of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  and to understand the origin of the strong mass enhancement observed in quantum oscillation and ARPES experiments in  $\text{KFe}_2\text{As}_2$ . Local density approximation + Dynamical mean field theory (LDA + DMFT) calculations stress that the mass enhancement in iron pnictides is not related to the proximity to a quantum critical point, but because the electrons, being rather localized at high temperature, start to

form coherent quasiparticle bands with the underlying Fermi surface [13,14]. In this scenario, Hund’s rule coupling is responsible for the large mass enhancement and a coherence-incoherence crossover is expected to occur for increasing temperature [13–15].

In this Letter, we combine resistivity, specific-heat, thermal-expansion, and susceptibility measurements to study in detail the normal state of  $\text{KFe}_2\text{As}_2$ . We clearly show that both the Sommerfeld coefficient ( $\gamma \approx 102 \text{ mJ mol}^{-1} \text{ K}^{-2}$ ) and the Pauli susceptibility ( $\chi \approx 4 \times 10^{-4}$ ) are strongly enhanced in this material. Further, our susceptibility and thermal-expansion measurements provide strong evidence for the existence of a crossover between a low-temperature heavy Fermi liquid and a high-temperature incoherent behavior reminiscent of heavy fermion physics. We use a Gutzwiller slave-boson mean-field analysis to obtain a more microscopic understanding of the mass enhancement. Our results reveal that  $\text{KFe}_2\text{As}_2$  is indeed close to an orbital-selective Mott transition (OSMT), which was predicted theoretically in Refs. [15–18], and that qualitatively explains the measured temperature dependences of the susceptibility and thermal expansion.

Figure 1 shows the low-temperature electrical resistivity  $\rho(T)$  of  $\text{KFe}_2\text{As}_2$  measured in the  $ab$  plane. A clear  $T^2$  dependence, with  $A \approx 0.025 \mu\Omega \text{ cm K}^{-2}$ , characteristic of a coherent Fermi liquid is observed. Although the resistive transition appears rather broad ( $\Delta T_c \approx 0.5 \text{ K}$ ), the estimated residual-resistivity ratio,  $\text{RRR} = \rho(300 \text{ K})/\rho(0 \text{ K}) \approx 1150$ , is surprisingly large, in agreement with previous reports [19,20]. Such seemingly

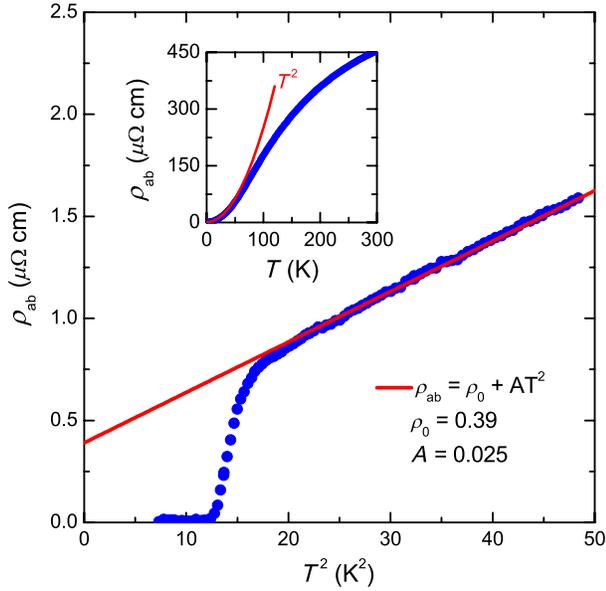


FIG. 1 (color online). Low-temperature electrical resistivity of  $\text{KFe}_2\text{As}_2$  as a function of  $T^2$ . The solid line is a fit to the usual Fermi liquid behavior,  $\rho_0 + AT^2$ . Units of  $\rho_0$  and  $A$  are  $\mu\Omega \text{ cm}$  and  $\mu\Omega \text{ cm K}^{-2}$ , respectively. The inset shows the temperature dependence of  $\rho(T)$  to 300 K.

contradictory behaviors suggest that the large RRR results not from an extremely low defect density [21], but rather from an anomalously high scattering rate at high temperature, as has been predicted by LDA + DMFT calculations with a Hund's coupling constant of about  $J_H \approx 0.35 \text{ eV}$  [13,14]. We note that a similar situation has already been pointed out for the heavy fermion superconductor  $\text{URu}_2\text{Si}_2$  [22]. The resistivity of  $\text{KFe}_2\text{As}_2$ , however, keeps increasing with temperature in contrast to heavy fermions for which a broad maximum is typically observed around the onset of coherence.

Figure 2 shows our sample heat capacity down to 0.2 K. A clear jump at  $T_c = 3.4 \text{ K}$ , with  $\Delta C/T_c \approx 54 \text{ mJ mol}^{-1} \text{ K}^{-2}$ , marks the transition to the bulk superconducting state. These values are in close agreement with previous results [10,23–25]. The excess heat capacity observed below 1 K and the modest normalized jump  $\Delta C/\gamma T_c \approx 0.53$  with respect to the BCS value ( $= 1.43$ ) are clear hallmarks of the existence of several energy gaps; the overall curve bears a strong similarity to that of  $\text{MgB}_2$  [26]. The solid line in Fig. 2 represents a fit to the normal-state heat capacity from which  $\gamma = 102 \text{ mJ mol}^{-1} \text{ K}^{-2}$  and the Debye temperature  $\Theta_D \approx 177 \text{ K}$  are determined, in agreement with previous reports [10,23]. As shown in the inset of Fig. 2, our value of  $\gamma$  complies with the requirement that the superconducting- and normal-state entropies are equal at  $T_c$ , excluding the existence of disordered magnetic phases reported in Ref. [25].  $\gamma$  is almost 9 times larger than the bare LDA value in  $\text{KFe}_2\text{As}_2$  while only a factor of about 2 is required to reconcile band-structure calculations with calorimetric

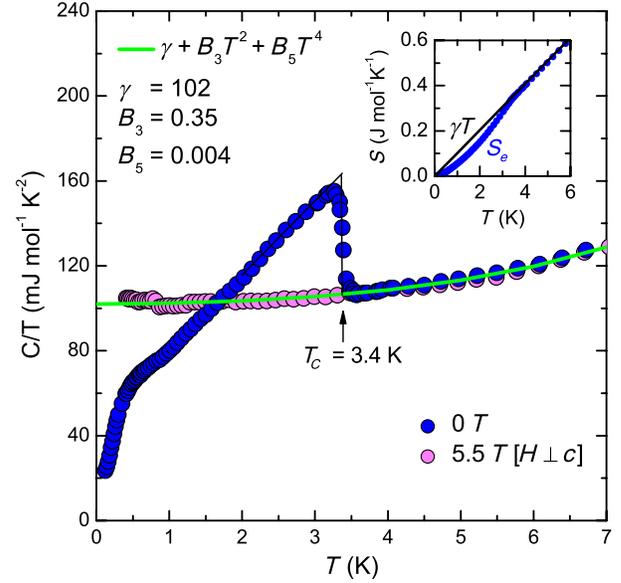


FIG. 2 (color online). Temperature dependence of the heat capacity of  $\text{KFe}_2\text{As}_2$  in 0 and 5.5 T. The green line represents a fit to the normal-state heat capacity. The Sommerfeld coefficient  $\gamma$ , the Debye term  $B_3$ , and  $B_5$  are given in  $\text{mJ mol}^{-1} \text{ K}^{-2}$ ,  $\text{mJ mol}^{-1} \text{ K}^{-4}$ , and  $\text{mJ mol}^{-1} \text{ K}^{-6}$ , respectively. The inset shows the normal- and superconducting-state electronic entropies.

measurements in  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$  for  $0 < x < 0.4$  [27]. The large Kadowaki-Woods ratio  $A/\gamma^2 \approx 2 \times 10^{-6} \mu\Omega \text{ cm K}^2 \text{ mol}^2 \text{ mJ}^{-2}$  clearly shows that  $\text{KFe}_2\text{As}_2$  is a highly correlated metal. Our value agrees well with the existence of heavy bands detected by de Haas-van Alphen and ARPES experiments, which estimate  $\gamma$  to be roughly  $82\text{--}94 \text{ mJ mol}^{-1} \text{ K}^{-2}$  [5–9]. Previously such unusually large  $\gamma$  values in  $d$  metals were found in  $\text{LiV}_2\text{O}_4$  and  $\text{Sr}_2\text{RuO}_4$ , for which  $\gamma$  reaches 420 and  $37.5 \text{ mJ mol}^{-1} \text{ K}^{-2}$ , respectively [28,29].

In strongly correlated systems, an enhanced  $\gamma$  with respect to the LDA value reflects the importance of correlations. It is often related to a characteristic temperature  $T^* \propto \gamma^{-1}$  which can be much lower than the bare electronic scale  $\epsilon_f/k_B$  (where  $\epsilon_f$  is the Fermi energy) because of these correlations. For  $T \ll T^*$ , quasiparticles are well defined, and the electronic heat capacity  $C_e$  is linear in temperature. For  $T \gg T^*$ , quasiparticles become short lived: the Landau Fermi-liquid description no longer applies and the heat capacity decreases with temperature to reach ultimately the undressed LDA value. For instance,  $T^*$  is referred to as the Kondo (or coherence) temperature in heavy fermion systems. Using susceptibility and dilatometry measurements, we show hereafter that this cross-over is clearly observed in  $\text{KFe}_2\text{As}_2$ .

Figure 3(a) shows the temperature dependence of the raw magnetic susceptibility  $M/H$  of  $\text{KFe}_2\text{As}_2$  measured for  $H \parallel c$  and  $H \perp c$  at several constant magnetic fields. For both orientations,  $M/H$  increases with decreasing temperatures and exhibits a broad maximum around

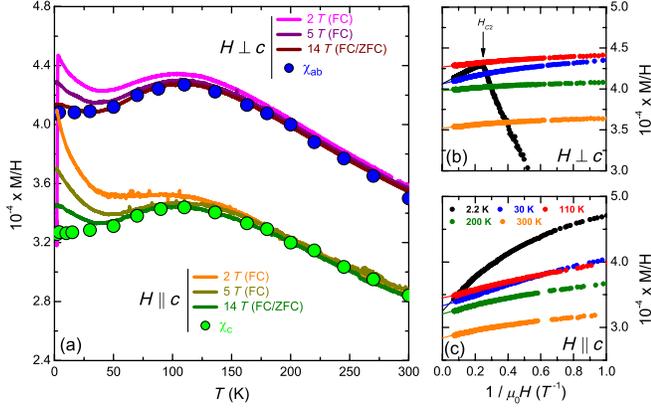


FIG. 3 (color online). (a) Measured magnetic susceptibility  $M/H$  of  $\text{KFe}_2\text{As}_2$  for  $H \perp c$  and  $H \parallel c$  for several magnetic fields (solid lines). The symbols show the temperature dependence of the intrinsic susceptibilities  $\chi_c(T)$  and  $\chi_{ab}(T)$  of  $\text{KFe}_2\text{As}_2$  derived from magnetic isotherms. (b),(c)  $M(H)|_T$  isotherm measurements performed at various temperatures, shown in a Honda-Owen representation.

100 K. At lower temperatures, a significant upturn, that vanishes with increasing field, develops. This upturn is typical of iron pnictide superconductors and reveals the presence of a small amount of magnetic or paramagnetic impurities, e.g., Fe atoms. Hence,  $M(T)|_H$  curves cannot be used to derive the intrinsic susceptibilities  $\chi_{ab}(T)$  and  $\chi_c(T)$  of  $\text{KFe}_2\text{As}_2$ . Alternatively, they can be determined precisely by analyzing magnetic isotherms,  $M(H)|_T$ , performed at various temperatures. As argued by Johnston [30], one has in this case:

$$M(H)|_T = \chi(T)H + M_i(H)|_T, \quad (1)$$

where  $\chi(T)$  and  $M_i(H)|_T$  are the intrinsic susceptibility of  $\text{KFe}_2\text{As}_2$  and the impurity contribution to  $M(H)|_T$ , respectively. Since  $M_i(H)|_T$  is not linear with field and typically saturates at some field,  $\chi(T)$  represents the slope of the high-field linear region of the measured  $M(H)|_T$  curves. Figures 3(b) and 3(c) show several  $M(H)|_T$  isotherms for both orientations displayed in a Honda-Owen representation [31,32] which gives the intrinsic  $\chi(T)$  by extrapolating  $M/H$  to  $1/H \rightarrow 0$ , according to Eq. (1). The derived intrinsic susceptibilities for  $H \parallel c$  and  $H \perp c$  are shown in Fig. 3(a) as solid circles. For  $T < 50$  K, both  $\chi_c$  and  $\chi_{ab}$  are nearly temperature independent and rather large, equal to  $3.2 \times 10^{-4}$  and  $4.1 \times 10^{-4}$ , respectively. This Pauli paramagnetic behavior is consistent with the Fermi liquid behavior observed in resistivity and heat-capacity measurements. With these values, we find a Wilson ratio  $R_W = \pi^2 k_B^2 V_m \chi / 3 \mu_0 \mu_B^2 \gamma$  of 1.1 and 1.3 for  $H \parallel c$  and  $H \perp c$ , respectively. A Wilson ratio close to unity indicates that the same quasiparticles are involved in the enhancement of both  $\gamma$  and the Pauli susceptibility. Moreover, the magnetic susceptibility is weakly anisotropic with a temperature-independent ratio  $\chi_{ab}/\chi_c \approx 1.3$ , which is in good

agreement with the Knight shift ratio,  $K_{ab}/K_c \approx 1.2-1.5$ , extracted from NMR measurements [33,34]. Finally, for  $T > 50$  K,  $\chi_{ab}$  and  $\chi_c$  increase and reach a broad maximum around 100 K and then monotonically decrease beyond, following approximately a Curie-Weiss law,  $\chi = C/(T + \Theta_{\text{CW}})$ , for  $T > 150$  K from which a fluctuating paramagnetic moment of about  $2.5 \mu_B$  is inferred and  $\Theta_{\text{CW}} \approx 600$  K which provides a crude estimation of  $T^*$ . This overall behavior is extremely similar to that of the heavy fermion  $\text{CeRu}_2\text{Si}_2$  which is paramagnetic but close to an antiferromagnetic instability [35].

In  $\text{CeRu}_2\text{Si}_2$ , the electronic heat capacity  $C_e/T$  is constant at low temperature with an enhanced  $\gamma = 360 \text{ mJ mol}^{-1} \text{ K}^{-2}$  and then decreases monotonically for  $T > 5$  K [36]. Thus, a similar behavior should be observed for  $\text{KFe}_2\text{As}_2$  but this crossover is complicated to address experimentally in calorimetric measurements because of the large phonon background. On the other hand, measurements of the coefficient of linear thermal expansion,

$$\alpha_i(T) = \frac{1}{L_i} \left( \frac{\partial L_i}{\partial T} \right)_{p_i} = - \left( \frac{\partial S}{\partial p_i} \right)_T \quad (i = a, c), \quad (2)$$

which probe the changes of entropy with respect to uniaxial pressure  $p_i$ , often show a relatively larger electronic signal compared to the heat capacity. This is particularly true for the iron-pnictide superconductors, which show large electronic signatures in the thermal expansion [27,37]. Figure 4 shows the in-plane thermal expansion of  $\text{KFe}_2\text{As}_2$  and, at low temperature, a clear Fermi-liquid behavior  $\alpha = aT$ , with a large electronic term  $a = 1.37 \times 10^{-7} \text{ K}^{-2}$  is observed. The raw data also clearly demonstrate that the low-temperature linear behavior does not survive up to higher temperatures, since the data near room temperature are actually decreasing with increasing temperature. In order to get a better measure of the electronic signal, we subtracted the thermal expansion of  $\text{Ba}(\text{Fe}_{0.67}\text{Co}_{0.33})_2\text{As}_2$ . This material was previously shown to have almost no electronic term [27] and, due to the similar crystal structure, should thus represent to a good approximation the phonon thermal expansion. The resulting electronic term,  $\alpha_e(T)$ , increases linearly at low temperatures, passes over a maximum near 125 K and then further decreases up to 300 K. This behavior clearly demonstrates the expected crossover from a low-temperature Fermi liquid regime to a high-temperature regime with a strongly reduced electronic thermal expansion. In Fig. 4(b), we show  $\alpha_e/T$ , whose temperature dependence bears a strong similarity to that of the susceptibilities shown in Fig. 3(a). Assuming  $C_e \propto \alpha_e$ , Fig. 4(b) also reflects the temperature variation of the electronic heat capacity  $C_e/T$ . Thus, our thermal-expansion and susceptibility measurements both show clear evidence of the coherence-incoherence crossover predicted by LDA + DMFT calculations [13]. Although there are clear similarities between  $\text{KFe}_2\text{As}_2$  and heavy fermions, the direct

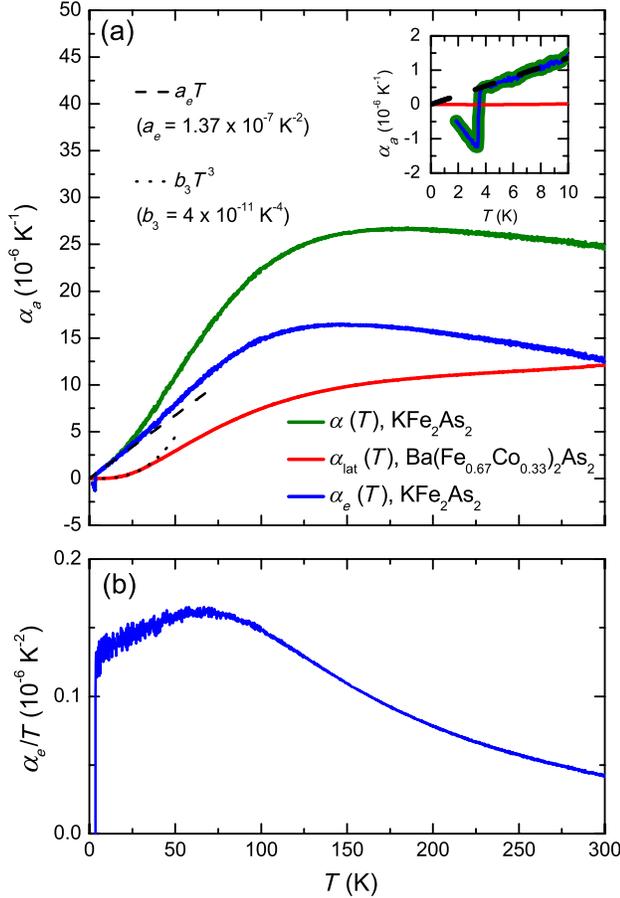


FIG. 4 (color online). (a) Temperature dependence of the thermal expansion of  $\text{KFe}_2\text{As}_2$  and  $\text{Ba}(\text{Fe}_{0.67}\text{Co}_{0.33})_2\text{As}_2$  measured along the  $a$  axis. The dashed and dotted lines are low-temperature fits to the Fermi liquid ( $aT$ ) and Debye ( $b_3T^3$ ) models, respectively. The blue line shows the electron thermal expansion of  $\text{KFe}_2\text{As}_2$  obtained by subtracting the expansivity of  $\text{Ba}(\text{Fe}_{0.67}\text{Co}_{0.33})_2\text{As}_2$  from that of  $\text{KFe}_2\text{As}_2$ . The inset shows a magnified view of the low-temperature regime. (b) Temperature dependence of  $\alpha_e/T$ .

comparison is probably too simplistic since there are no  $4f$  electrons involved in this iron pnictide. Nevertheless, our measurements clearly illustrate the existence of strong antiferromagnetic correlations in  $\text{KFe}_2\text{As}_2$ . Beside large  $\gamma$  values, similar temperature dependences were reported in other correlated  $d$ -metal alloys, e.g., the layer perovskite  $\text{Ca}_{2-x}\text{Sr}_x\text{RuO}_4$  [38]. In these materials, the origin of the heavy-fermion-like behavior is still unsettled. But theoretical calculations (DMFT + LDA) ascribe it to an OSMT since hybridization induces orbital fluctuations that results in the formation of a Kondo-like heavy-fermion behavior [39,40]. The proximity to such an OSMT is actually debated in iron pnictide superconductors.

In order to obtain a more microscopic understanding of the origin of the significant mass enhancement in  $\text{KFe}_2\text{As}_2$ , we performed a Gutzwiller corrected electronic structure calculation following our earlier analysis for related

systems [17]. Using a tight-binding fit of the iron  $3d$  bands, we include local intra- and interorbital Coulomb repulsions  $U$  and  $U'$ , respectively, as well as Hund's coupling  $J_H$  between the various electrons. In order to describe and characterize electronic correlation effects we performed a Gutzwiller slave-boson mean-field calculation that yields orbitally resolved mass enhancement factors  $z_\alpha$  where  $\alpha$  refers to the five  $d$  orbitals. Our results are shown in Fig. 5 where we plot  $z_\alpha(J_H)$  for different values of  $U$  for  $\text{KFe}_2\text{As}_2$  and  $\text{BaFe}_2\text{As}_2$ . In both materials, the Fermi surface sheets of  $|xy\rangle$  character are the most strongly coupled, followed by  $|xz\rangle$ ,  $|yz\rangle$ , and  $|z^2\rangle$  states. These overall trends are similar to what is found in LDA-DMFT calculations [14]. One important distinction between  $\text{BaFe}_2\text{As}_2$  and  $\text{KFe}_2\text{As}_2$  is clearly the more prominent role played by  $|xy\rangle$  states on the Fermi surface. We also find a significant difference in the values of the effective masses  $m^* \propto z_\alpha^{-1}$  for  $\text{KFe}_2\text{As}_2$  in comparison with  $\text{BaFe}_2\text{As}_2$  if we consider sufficiently large values of the local Coulomb interaction and the Hund's coupling. For  $U \geq 4$  eV we find for  $J_H \geq 0.5$ – $0.6$  eV an orbitally selective Mott transition with a localization of the  $|xy\rangle$  states. While these parameters are too large for a realistic description of the material, they demonstrate the proximity of  $\text{KFe}_2\text{As}_2$  to an OSMT for the configuration intermediate between  $3d^5$  and  $3d^6$ , a concept that was already discussed in the pnictide family in Refs. [15–18]. The fact that the significant mass enhancement occurs for  $|xy\rangle$  dominated bands is in complete agreement with quantum oscillation [5,6,9] and ARPES experiments [7,8]. At the actual Mott transition, correlated electron states lose their metallic character and localize due to the strong Coulomb repulsion. In the case of an OSMT, this localization only occurs for some of the involved

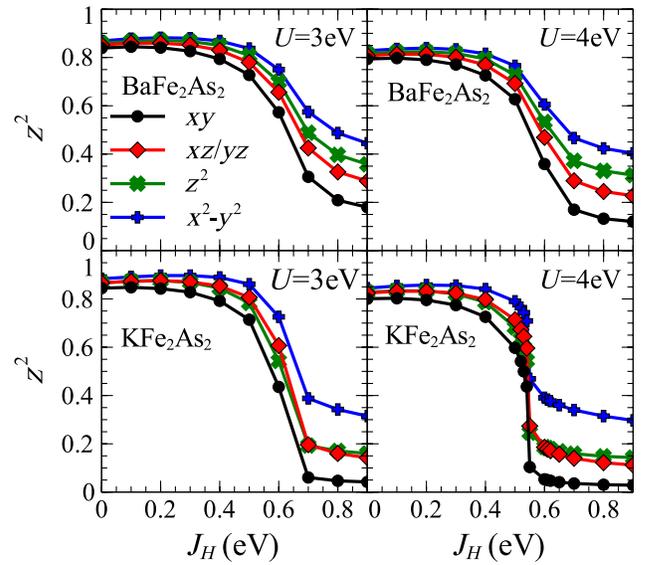


FIG. 5 (color online). Gutzwiller slave-boson mean-field calculations of the orbitally resolved mass-enhancement factor  $z_\alpha$  for  $\text{BaFe}_2\text{As}_2$  and  $\text{KFe}_2\text{As}_2$ , for two different values of the intraorbital Coulomb repulsion  $U$ .

orbitals, while others remain itinerant [15,16,40], as can be seen from the orbital dependence of the quasiparticle weight shown in Fig. 5. This dichotomy of localized and delocalized bands can then give rise to a behavior very similar to that of  $4f$ -based heavy-electron systems, even though all states are formed from Fe  $3d$  orbitals, as seen in recent theoretical and experimental studies in other iron pnictides [41,42].

In summary, by studying the normal-state properties of  $\text{KFe}_2\text{As}_2$ , we have shown that it is a strongly correlated material with highly renormalized values of both the Sommerfeld coefficient and the Pauli susceptibility. Additionally, we have provided evidence for a temperature-induced incoherent-coherent crossover to this highly correlated state. The physics of these correlations and this crossover are well described by the possible close proximity of  $\text{KFe}_2\text{As}_2$  to an orbital-selective Mott transition due to Hund's coupling between orbitals. It is interesting to note that these strong correlations do not enhance superconductivity— $T_c$  is rather low and does not correlate with  $\gamma$  for different K dopings. In fact, the correlations appear to be detrimental to superconductivity, which may be a rather general feature of high-temperature superconductivity, e.g., in the 115 class of compounds, the highest  $T_c \approx 18$  K is found for the weakly correlated Pu compound [43] while  $\text{CeIrIn}_5$ , with a large  $\gamma \approx 800$  mJ mol $^{-1}$  K $^{-2}$ , shows a quite low  $T_c$  [44].

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