Spin excitations in optimally P-doped BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ superconductor

Ding Hu,1,2 Zhiping Yin,1,3,∗ Wenliang Zhang,2 R. A. Ewings,4 Kazuhiko Ikeuchi,5 Mitsutaka Nakamura,6 Bertrand Roessli,7 Yuan Wei,3 Lingxiao Zhao,2 Genfu Chen,2 Shiliang Li,2,8 Huiqian Luo,2 Kristjan Haule,3 Gabriel Kotliar,3,9 and Pengcheng Dai10,1,

1Center for Advanced Quantum Studies and Department of Physics, Beijing Normal University, Beijing 100875, China
2Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China
3Department of Physics, Rutgers University, Piscataway, New Jersey 08854, USA
4ISIS Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot OX11 0QX, United Kingdom
5Research Center for Neutron Science and Technology, Comprehensive Research Organization for Science and Society (CROSS), Tokai, Ibaraki 319-1106, Japan
6Materials and Life Science Division, J-PARC Center, Tokai, Ibaraki 319-1195, Japan
7Laboratory for Neutron Scattering and Imaging, Paul Scherrer Institut, CH-5232 Villigen, Switzerland
8Collaborative Innovation Center of Quantum Matter, Beijing, China
9Brookhaven National Laboratory, Upton, New York 11973-5000, USA
10Department of Physics and Astronomy, Rice University, Houston, Texas 77005, USA

(Received 13 February 2016; revised manuscript received 26 July 2016; published 2 September 2016)

We use inelastic neutron scattering to study the temperature and energy dependence of spin excitations in an optimally P-doped BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ superconductor ($T_c = 30$ K) throughout the Brillouin zone. In the undoped state, spin waves and paramagnetic spin excitations of BaFe$_2$As$_2$ stem from an antiferromagnetic (AF) ordering wave vector $Q_{\text{AF}} = (\pm 1, 0)$, and peak near the zone boundary at ($\pm 1, \pm 1$) around 180 meV. Replacing 30% As by smaller P to induce superconductivity, low-energy spin excitations of BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ form a resonance in the superconducting state and high-energy spin excitations now peak around 220 meV near ($\pm 1, \pm 1$). These results are consistent with calculations from a combined density functional theory and dynamical mean field theory, and suggest that the decreased average pnictogen height in BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ reduces the strength of electron correlations and increases the effective bandwidth of magnetic excitations.

DOI: 10.1103/PhysRevB.94.094504

I. INTRODUCTION

Since the discovery of unconventional superconductivity in iron pnictides near antiferromagnetic (AF) instability [1–7], a central issue has been whether these materials are fundamentally different from copper oxide superconductors, where the magnetism and superconductivity are derived from Mott physics and its associated electron correlations [8–14]. Since iron pnictides have tetrahedrally coordinated nearest pnictogen atoms, the 3$d$ level in Fe ions splits into an $e_g$ state and a few hundred meV higher $t_{2g}$ state [15–22]. Without Hund’s rule interaction, the $e_g$ state would be fully occupied with four of the six Fe 3$d$ electrons while the remaining two 3$d$ electrons should reside in the $t_{2g}$ state crossing the Fermi level. The presence of a strong Hund’s coupling $J_H$, which tends to align spins of all the electrons on a given Fe atom, competes with the crystal field splitting and promotes high spin states of the Fe 3$d$ electrons, resulting in large charge and spin fluctuations. Although it is generally accepted that electron correlations are also present in iron pnictides [11–14], it remains unclear if the correlation strength is controlled by the on-site Hubbard $U$ interaction as in the case of cuprates [8] or if it arises primarily from the Hund’s coupling $J_H$ within one atomic site [15–17]. The local moments formed by the Fe 3$d$ electrons, especially those in the $d_{xz}$, $d_{yz}$, and $d_{xy}$ orbitals, are coupled to their nearest neighbors by both the direct exchange associated with nearest-neighbor Fe-Fe distance and anisotropic superexchange interactions via hopping through the As pnictogen [Figs. 1(a)–1(c)]. By increasing the Fe-pnictogen distance, electron hopping between the nearest Fe ions becomes difficult, and the system is localized with enhanced electron correlations. On the other hand, reducing the Fe-pnictogen distance facilitates the electron hopping, thus reducing the electron correlations.

Using density functional theory (DFT) combined with dynamical mean field theory (DMFT) suitable for describing the Hund’s coupling in iron pnictides [15–17,23,24], the evolution of spin excitations in electron- and hole-doped BaFe$_2$As$_2$ can be calculated [25–27]. In particular, the theory predicts that spin-wave and spin excitation bandwidths, defined as the peak in energy dependence of the dynamic susceptibility $\chi''(E)$ integrated near the AF zone boundary ($\pm 1, \pm 1$) [25,27], in different iron pnictides are controlled by the iron-pnictogen distance and the valence of the Fe atoms similar to electron correlations [28]. Experimentally, spin waves and spin excitations in the electron- and hole-doped AFe$_2$As$_2$ (A = Ba, Sr, Ca) family of iron pnictides mapped out by inelastic neutron scattering experiments throughout the Brillouin zone [29–33] are qualitatively consistent with the DFT+DMFT calculations and peak around 180 meV near (1, 1) [25,27]. Although neutron scattering experiments on spin waves of NaFeAs, which has a larger pnictogen height ($h_{Pn} = 1.416$ Å) [34] compared with that of BaFe$_2$As$_2$ [Fig. 1(b), $h_{Pn} = 0.36$ Å], confirm the notion that increasing pnictogen height in iron pnictides decreases the spin-wave bandwidths to $\sim 110$ meV near (1, 1) and increases the electron correlation...
[35], the crystal structures of NaFeAs and BaFe$_2$As$_2$ are rather different, and it is still unclear that varying the iron-pnictogen distance within one family of iron pnictides can indeed control the electron correlations and spin excitation spectra.

In this paper, we present inelastic neutron scattering studies of the temperature and energy dependence of spin excitations in a BaFe$_2$(As$_{0.8}$P$_{0.2}$)$_2$ superconductor ($T_c = 30$ K) [36]. We chose BaFe$_2$(As$_{0.8}$P$_{0.2}$)$_2$ because it is the optimal isovalently doped BaFe$_2$As$_2$. At the same time, it has an average pnictogen height ($h_{\text{As}} = 1.28$ Å) significantly smaller than that of BaFe$_2$As$_2$ due to the smaller size of the P dopants [Figs. 1(b) and 1(c)] [4]. Since the average pnictogen height of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ decreases continuously from BaFe$_2$As$_2$ to BaFe$_2$(As$_{0.8}$P$_{0.2}$)$_2$ without modifying much the in-plane Fe-Fe distance or changing the valence of Fe [4], we expect weaker electron correlations and a wider spin excitation bandwidth in BaFe$_2$(As$_{0.8}$P$_{0.2}$)$_2$ compared with that of BaFe$_2$As$_2$ [28].

Figures 1(d) and 1(e) show the energy dependence of $S(Q,E)$ of spin excitations for BaFe$_2$As$_2$ and BaFe$_2$(As$_{0.8}$P$_{0.2}$)$_2$, respectively. While spin-wave dispersions of BaFe$_2$As$_2$ reach zone boundary positions (1,±1) around 200 meV [Fig. 1(d)], dispersions of spin excitations of BaFe$_2$(As$_{0.8}$P$_{0.2}$)$_2$ become steeper, and reach (1,±1) at energies well above 200 meV [Fig. 1(e)]. Indeed, we find that spin excitations in BaFe$_2$(As$_{0.8}$P$_{0.2}$)$_2$ have a lower intensity but a larger energy bandwidth than that of BaFe$_2$As$_2$ [30], as revealed in the energy dependence of the dynamic susceptibility $\chi''(E)$ integrated over the dashed vertical lines in Figs. 1(d) and 1(e) ($0.6 \leq H \leq 1.4$ and $0.6 \leq K \leq 1.4$) near the zone boundary (1,±1) [Figs. 1(f) and 1(g)]. These results are also consistent with the energy dependence of the local dynamic susceptibility and DMFT calculations of $S(Q,E)$ [Figs. 1(h) and 2–4]. Therefore, the decreased average pnictogen height in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ decreases the electron correlations and increases the overall spin excitation energy bandwidths.

II. EXPERIMENTAL AND THEORETICAL RESULTS

Our neutron scattering experiments were carried out on the MAPS and 4SEASONS chopper spectrometers at ISIS, Rutherford Appleton Laboratory, U.K. and Japan Proton Accelerator Research Complex, Japan, respectively. Some measurements are also carried out on the EIGER triple-axis spectrometer at the Paul Scherrer Institut, Switzerland. Our BaFe$_2$(As$_{0.8}$P$_{0.2}$)$_2$ single crystals were grown by a self-flux method [36]. For P-doped BaFe$_2$(As$_{1-x}$P$_x$)$_2$ near $x = 0.3$, the collinear static AF order in BaFe$_2$As$_2$ is suppressed and superconductivity reaches an optimal value at $T_c = 30$ K [37]. We coiled a few g of single crystals in the [H,H,L] scattering plane with a mosaic <7'. To facilitate easy comparison with spin waves in BaFe$_2$As$_2$, which has an orthorhombic AF ground state [30], we define the wave vector $Q$ at

![FIG. 1. (a) The crystal structure of BaFe$_2$(As$_{1-x}$P$_x$)$_2$. The purple, silver, and blue balls indicate Fe, As/P, and Ba positions, respectively. (b), (c) Schematic diagrams of the FeAs tetrahedron, showing the average iron-pnictogen height decreased from 1.36 Å for BaFe$_2$As$_2$ to 1.28 Å for BaFe$_2$(As$_{0.8}$P$_{0.2}$)$_2$ [37]. (d) The energy dependence of $S(Q,E)$ of spin waves of BaFe$_2$As$_2$ along the (1, K) direction (with integration of $H$ from 0.9 to 1.1 r.l.u.) after subtracting the background integrated from 1.8 < $H$ < 2.2 and from −0.25 < $K$ < 0.25 r.l.u. with $E_i = 450$ meV at $T = 10$ K measured on MAPS [30]. (e) Identical projection for spin excitations of BaFe$_2$(As$_{0.8}$P$_{0.2}$)$_2$ obtained on MAPS with $E_i = 450$ meV. The negative scattering below ~50 meV is due to errors in background subtraction. Energy dependence of wave vector integrated [integration range is shown in the shaded box in the inset of (g)] dynamic susceptibility $\chi''(E)$ for (f) BaFe$_2$As$_2$ and (g) BaFe$_2$(As$_{0.8}$P$_{0.2}$)$_2$. The vertical dashed lines in (d) and (e) show the wave vector integration range along the [0,K] direction. (h) Energy dependence of the local dynamic spin susceptibility $\chi''(E)$ for BaFe$_2$As$_2$ (solid cyan line) and BaFe$_2$(As$_{0.8}$P$_{0.2}$)$_2$ below (solid red triangle and solid red line)
energies were inelastic neutron scattering measurements, the incident beam weaker and more diffusive in the momentum space in P doping, the hole Fermi surface with a dominating 3) of optimally P-doped BaFe2(As1−xP)x, occurring at 1.28 Å in the BaFe2P2.

Since there is no evidence that the P dopant forms long-range order in BaFe2(As1−xP)x, we use an effective pnictide position in the DFT+DMFT calculations to simulate the physical consequence of P doping. In BaFe2(As0.7P0.3)2, the As and P heights are 1.332 and 1.151 Å from the Fe plane, respectively [38]. We therefore take an effective pnicogen height of 1.28 Å in our calculation, which is the average height of As and P in BaFe2(As0.7P0.3)2 determined experimentally. This effective As/P height (1.28 Å) is less than the As height (1.36 Å) in the BaFe2As2 but substantially larger than the P height (1.19 Å) in the BaFe2P2.

In previous inelastic neutron scattering studies of low-energy spin excitations in powder [39] and single crystals [40] of optimally P-doped BaFe2(As1−xP)x, a neutron spin resonance coupled to superconductivity has been identified similar to other iron-based superconductors [41–43]. A key conclusion of the work is that the energy of the resonance in BaFe2(As0.63P0.37)2 is dispersive along the c axis, indicating its close connection to the three-dimensional AF spin correlations [40]. In hole- and electron-doped BaFe2As2, the wave vector evolution of the low-energy spin excitations and the resonance can be well described by quasiparticle excitations through doping-dependent hole and electron Fermi surfaces [27,44]. Although substituting P for As in BaFe2(As1−xP)x is expected to be isovalent, angle-resolved photoemission spectroscopy (ARPES) experiments reveal changed hole and electron Fermi surfaces from BaFe2As2 to BaFe2(As0.5P0.5)2 [45]. DFT+DMFT calculations also show that with increasing P doping, the hole Fermi surface with a dominating dxz orbital character shrinks whereas the hole Fermi surfaces with dominating dz2 and dxz (dz2 near kz = π) orbital characters expand and become more three dimensional along the kz direction [Figs. 2(a) and 2(b)]. However, the electron Fermi surfaces do not change significantly. Therefore, the electron-hole Fermi surface nesting condition becomes worse with P doping. This is consistent with the resulting changes in the wave vector dependence of spin excitations [Figs. 2(c) and 2(d)], where the low-energy spin excitations become weaker and more diffuse in the momentum space in BaFe2(As0.7P0.3)2. Figures 2(e) and 2(f) show constant-Q scans at Q = (1,0,0) and (1,0,1), respectively, below and above Tc. Consistent with previous work [40], we find that superconductivity-induced resonance is clearly dispersive, occurring at Eres = 12 meV at Q = (1,0,0) and Eres = 9 meV at Q = (1,0,1). Figures 2(g) and 2(h) summarize the energy dependence of the low-energy spin excitations at 40 K (T ≈ Tc + 10 K) and 10 K (T ≈ Tc − 20 K), respectively. Given the dispersive nature of the resonance, neutron time-of-flight measurements with a fixed incident energy and fixed sample rotation angle in Figs. 2(g) and 2(h) will probe a region of the excitation energies with different L values. Figure 2(i) is a temperature difference plot, revealing a clear neutron spin resonance in the energy region of E ≈ 11 meV and a spin gap below the resonance energy.
Assuming that isovalent P doping in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ does not change the valence of Fe, the total moment sum rule requires the total magnetic spectral weight $M_0$, when integrated over all energy and momentum space $[M_0^2 = M^2 + (\textbf{m}_f^2)] = g^2S(S + 1)$, where $M$ is the static ordered moment, $(\textbf{m}_f^2)$ is the local fluctuating moment, $g \approx 2$ is the Landé factor, and $S$ is the spin), to be independent of P doping [7]. From Figs. 2(c) and 2(d), we see a reduced low-energy spin excitation spectral weight in BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ compared with BaFe$_2$As$_2$, as revealed by the DFT+DMFT calculation [Figs. 1(h) and 4].

Figures 3(a)–3(e) and 3(k)–3(o) compare the two-dimensional constant-energy $(E)$ images of $S(Q,E)$ of spin waves of BaFe$_2$As$_2$ [30] and spin excitations of BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ in the $(H,K)$ scattering plane at different energies. Figures 3(a)–3(e) show the evolution of spin waves of BaFe$_2$As$_2$ at energy transfers of $E = 30 \pm 5$, $50 \pm 5$, $95 \pm 5$, $157 \pm 10$, and $225 \pm 10$ meV, respectively. The corresponding spin excitations of BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ are shown in Figs. 3(k)–3(o). At $E = 30 \pm 5$ [Figs. 3(a) and 3(k)] and $E = 50 \pm 5$ meV [Figs. 3(b) and 3(l)], spin excitations in BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ form transversely elongated ellipses centered at the in-plane AF zone centers $(\pm 1,0)$ and $(0,\pm 1)$ of the undoped BaFe$_2$As$_2$, but with considerably lower intensity. On increasing the energies to $E = 95 \pm 5$ meV [Fig. 3(m)], spin excitations of BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ begin to split transversely from $(\pm 1,0)$, similar to that of spin waves of BaFe$_2$As$_2$ [Fig. 3(c)]. On further increasing the energy to $E = 157 \pm 10$ [Fig. 3(n)] and $E = 225 \pm 10$ meV [Fig. 3(o)], spin excitations of BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ form anisotropic rings centered around $(\pm 1,\pm 1)$. Figures 3(d) and 3(e) show spin waves of BaFe$_2$As$_2$ at $E = 157 \pm 10$ and $E = 225 \pm 10$ meV, respectively. We see that spin waves of BaFe$_2$As$_2$ at $E = 225 \pm 10$ meV nearly form a solid spot at $(\pm 1,\pm 1)$, suggesting that the system has

\[ x = 0 \text{ DFT + DMFT} \]

\[ x = 0 \text{ DFT} + \text{DMFT} \]
already reached the zone boundary at this energy. For comparison, spin excitations of BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ at $E = 225 \pm 10$ meV still have a ring structure near $(\pm 1, \pm 1)$ [Fig. 3(o)], as confirmed by a comparison of constant-energy cuts across the data (see Supplemental Fig. 5 in Ref. [46]). These results suggest a higher zone boundary energy for BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$.

To quantitatively compare the experimental results with a combined DFT+DMFT theory [28], we show in Figs. 3(f)–3(j) and 3(p)–3(t) the calculated wave vector dependence of spin excitations of BaFe$_2$As$_2$ and BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$, respectively, at energies in Figs. 3(a)–3(e) [28]. We see that spin excitations at different energies obtained from the DFT+DMFT calculation in Figs. 3(p)–3(t) have many similarities with the experimental data [Figs. 3(k)–3(o)].

Figures 4(a) and 4(b) show the calculated dynamical magnetic structure factor $S(Q,E)$ for BaFe$_2$As$_2$ and BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$, respectively. Our calculation reveals a considerable magnetic spectral weight for energies above 300 meV for both samples, contrasting to the vanishing local dynamical susceptibility for energies above 300 meV in BaFe$_2$As$_2$ [Fig. 1(h)] [47]. The experimentally determined spin excitation dispersion along the $[1,0]$ direction for BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ is well captured by the DFT+DMFT calculations [Fig. 4(b)]. For comparison, we also plot in Fig. 4(b) the experimentally determined dispersion along the same direction for BaFe$_2$As$_2$ as a solid line [30]. To understand the different orbital contributions to the spin excitations, we show in Figs. 4(c) and 4(d) the $d_{x^2-y^2}$ and $d_{z^2}$ intraorbital contributions to the dynamic susceptibility. We see that low-energy spin excitations near $(1,0)$ are mostly contributed by excitations involving the $d_{x^2}$ orbital, while high-energy spin excitations around 300 meV near $(1,1)$ come mostly from excitations related to the $d_{z^2}$ orbital. This is in stark contrast with spin excitations in the Co-doped LiFeAs compound, where the low-energy spin excitations are dominated by contributions from the $d_{z^2}$ orbital [48]. In Ref. [48], it was concluded that the $d_{x^2}$ or $d_{z^2}$ orbitals play an important role in the superconductivity of LiFeAs since their absence in LiFe$_{0.8}$Co$_{0.2}$As suppresses superconductivity. The strong low-energy spin excitations contributed by the $d_{x^2}$ orbitals in BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ shown in Fig. 4(d) suggest that the nesting of the $d_{z^2}$ orbitals is good for high-temperature superconductivity.

Figure 1(h) compares the energy dependence of the local dynamic spin susceptibility for BaFe$_2$As$_2$, BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$, and DFT+DMFT calculated values. We see that the peak for local dynamic spin susceptibility for BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ occurs around 220 meV, while it is around 180 meV for BaFe$_2$As$_2$. These results are consistent with energy cuts near the zone boundary for these materials shown in Figs. 1(f) and 1(g). The total fluctuating magnetic moments for BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ and BaFe$_2$As$_2$ are $(m^2) \approx 1.6 \pm 0.2$ (below $\approx 250$ meV) and 3.6$\mu_B$ per Fe, respectively [47]. This means that the fluctuating moments of BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ are smaller than those of BaFe$_2$As$_2$ within our energy integration region, consistent with the presence of more magnetic spectral weight at higher energies or a reduced fluctuating moment. These results thus suggest that the decreased iron-pnictogen height in iron pnictides from BaFe$_2$As$_2$ to BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ increases the spin excitation bandwidth and decreases the electron correlation effects. Since the reduced pnictogen height due to P doping induces indirect hopping between Fe 3$d$ orbitals and pnictogen $p$ orbitals, and weakens the kinetic frustration as direct hopping between Fe 3$d$ orbitals remains almost the same as the Fe-Fe distance changes negligibly with P doping, the bandwihds of the Fe 3$d$ orbitals increase with increasing P doping [46] and the electronic correlation effects decrease. The increased bandwidhs lead to a reduction of the one-particle Green’s function, and thus a reduction in the bare two-particle susceptibility. Neglecting the change in the two-particle vertex function due to P doping, this reduction in the bare two-particle susceptibility is responsible for the reduced low-energy spin excitation intensity. Similarly, the spin excitation bandwidths increase due to the reduced pnictogen height in BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$.

The reduction of the pnictogen height in BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ from BaFe$_2$As$_2$ reduces the low-energy spin excitation intensity centered at $Q_{AF}$ and eliminates the static long-range AF order in the undoped BaFe$_2$As$_2$. At the same time, the pnictogen height in BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ is still high enough to maintain an intermediate electronic correlation strength with sufficient low-energy spin fluctuations to mediate superconductivity. For spin excitation mediated superconductors [5], superconductivity is controlled by the effective magnetic exchange coupling $J$ and the strength of the electron-spin excitation coupling [27]. Since the effective magnetic exchange couplings in BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ are considerably larger than those of BaFe$_2$As$_2$, it would be interesting to compare superconductivity-induced changes in spin excitations...
of BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$ and electron/hole-doped BaFe$_2$As$_2$ [27]. By comparing the absolute intensity changes of the resonance below and above $T_c$, we find that spin excitations changes across $T_c$ are still much larger than the superconducting condensation energy [46,49], thus supporting the notion that magnetism is crucial for the superconductivity of BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$.

III. CONCLUSIONS

In summary, we have used inelastic neutron scattering to map out spin excitations of isovalently doped BaFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$. By comparing spin excitations of this material with those of BaFe$_2$As$_2$ and DFT+DMFT calculations, we conclude that the iron-pnictogen height in iron pnictides directly controls the spin excitation bandwidth and electron correlations. These results are consistent with the idea that electron correlations in iron-based superconductors arise primarily from the Hund’s coupling $J_H$, and low-energy spin excitations are consequences of nesting between hole and electron Fermi surfaces.

ACKNOWLEDGMENTS

The work at IOP, CAS is supported by National Natural Science Foundation of China (NSFC Projects: 11374011, 11374346, 11674406 and 11611130165), the Ministry of Science and Technology of China (973 projects: 2012CB821400 and 2015CB921302), and the Strategic Priority Research Program (B) of CAS (Grant No. XDB07020300). H. Luo is grateful for the support from the Youth Innovation Promotion Association, CAS (No. 2016004). Neutron scattering work at Rice is supported by the U.S. DOE, Office of Basic Energy Sciences, under Contract No. DE-SC0012311. Part of the materials work at Rice University is supported by the Robert A. Welch Foundation Grant No. C-1839. The neutron experiment at the Materials and Life Science Experimental Facility of J-PARC was performed under a user program (Proposal No. 2014B0277). The computational works at Rice and Rutgers are supported by NSF DMREF DMR-1436006 and DMR-1435918, respectively. Experiments at the ISIS Pulsed Neutron and Muon Source were supported by a beam time allocation from the Science and Technology Facilities Council.

[1] Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, J. Am. Chem. Soc. 130, 3296 (2008).
[2] C. de la Cruz, Q. Huang, J. W. Lynn, J. Li, W. Ratcliff, II, J. L. Zarestky, H. A. Mook, G. F. Chen, J. L. Luo, N. L. Wang, and P. C. Dai, Nature (London) 453, 899 (2008).
[3] S. Jiang, H. Xing, G. Xuan, C. Wang, Z. Ren, C. Feng, J. Dai, Z. Xu, and G. Cao, J. Phys.: Condens. Matter 21, 382203 (2009).
[4] D. C. Johnston, Adv. Phys. 59, 803 (2010).
[5] D. J. Scalapino, Rev. Mod. Phys. 84, 1383 (2012).
[6] T. Shibatachi, A. Carrington, and Y. Matsuda, Annu. Rev. Condens. Matter Phys. 5, 113 (2014).
[7] P. C. Dai, Rev. Mod. Phys. 87, 855 (2015).
[8] P. A. Lee, N. Nagaosa, and X.-G. Wen, Rev. Mod. Phys. 78, 17 (2006).
[9] J. Hirschfeld, M. M. Korshunov, and I. I. Mazin, Rep. Prog. Phys. 74, 124508 (2011).
[10] A. Chubukov, Annu. Rev. Condens. Matter Phys. 3, 57 (2012).
[11] Q. Si and E. Abrahams, Phys. Rev. Lett. 101, 076401 (2008).
[12] C. Fang, H. Yao, W. F. Tsai, J. P. Hu, and S. A. Kivelson, Phys. Rev. B 77, 224509 (2008).
[13] C. Xu, M. Muller, and S. Sachdev, Phys. Rev. B 78, 020501(R) (2008).
[14] D. N. Basov and A. V. Chubukov, Nat. Phys. 7, 272 (2011).
[15] K. Haule and G. Kotliar, New J. Phys. 11, 025021 (2009).
[16] Z. P. Yin, K. Haule, and G. Kotliar, Nat. Mater. 10, 932 (2011).
[17] A. Goerges, L. de’ Medici, and J. Mrrvlje, Annu. Rev. Condens. Matter Phys. 4, 137 (2013).
[18] C. C. Lee, W. G. Yin, and W. Ku, Phys. Rev. Lett. 103, 267001 (2009).
[19] F. Krüger, S. Kumar, J. Zaanen, and J. van den Brink, Phys. Rev. B 79, 054504 (2009).
[20] W. C. Lv, J. S. Wu, and P. Phillips, Phys. Rev. B 80, 224506 (2009).
[21] C.-C. Chen, J. Macejko, A. P. Sorini, B. Moritz, R. R. P. Singh, and T. P. Devereaux, Phys. Rev. B 82, 100504(R) (2010).
[22] B. Valenzuela, E. Bascones, and M. J. Calderón, Phys. Rev. Lett. 105, 207202 (2010).
[23] G. Kothe, S. Y. Savrasov, K. Haule, V. S. Oudovenko, O. Parcollet, and C. A. Marianetti, Rev. Mod. Phys. 78, 865 (2006).
[24] K. Haule, C.-H. Yee, and K. Kim, Phys. Rev. B 81, 195107 (2010).
[25] M. S. Liu, W. Harriger, H. Q. Luo, M. Wang, R. A. Ewing, T. Guidi, H. Park, K. Haule, G. Kotliar, S. M. Hayden, and P. C. Dai, Nat. Phys. 8, 376 (2012).
[26] H. Park, K. Haule, and G. Kotliar, Phys. Rev. Lett. 107, 137007 (2011).
[27] M. Wang, C. L. Zhang, X. Y. Lu, G. T. Tan, H. Q. Luo, Y. Song, M. Y. Wang, X. T. Zhang, E. A. Goremeykin, T. G. Perring, T. A. Maier, Z. P. Yin, K. Haule, G. Kotliar, and P. C. Dai, Nat. Commun. 4, 2874 (2013).
[28] Z. P. Yin, K. Haule, and G. Kotliar, Nat. Phys. 10, 845 (2014).
[29] S. O. Diallo, V. P. Antropov, T. G. Perring, C. Broholm, J. A. Pulsikutit, N. Ni, S. L. Bud’ko, P. C. Canfield, A. Kreyssig, A. I. Goldman, and R. J. McQueeney, Phys. Rev. Lett. 102, 187206 (2009).
[30] L. W. Harriger, H. Q. Luo, M. S. Liu, C. Frost, J. P. Hu, M. R. Norman, and P. C. Dai, Phys. Rev. B 84, 054544 (2011).
[31] R. A. Ewing, T. G. Perring, J. Gillett, S. D. Das, S. E. Sebastian, A. E. Taylor, T. Guidi, and A. T. Boothroyd, Phys. Rev. B 83, 214519 (2011).
[32] J. T. Park, G. Fimmel, T. Loew, V. Hinkov, Yuan Li, B. H. Min, D. L. Sun, A. Ivanov, A. Piovano, C. T. Lin, B. Keimer, Y. S. Kwon, and D. S. Inosov, Phys. Rev. B 86, 024437 (2012).
[33] H. Q. Luo, X. Y. Lu, R. Zhang, M. Wang, E. A. Goremeykin, D. T. Adroja, S. Danilkin, G. Deng, Z. Yamani, and P. C. Dai, Phys. Rev. B 88, 144516 (2013).
[34] S. L. Li, C. de la Cruz, Q. Huang, G. F. Chen, T.-L. Xia, J. L. Luo, N. L. Wang, and P. C. Dai, Phys. Rev. B 80, 020504(R) (2009).
[35] C. L. Zhang, L. W. Harriger, Z. P. Yin, W. C. Lv, M. Y. Wang, G. T. Tan, Y. Song, D. L. Abernathy, W. Tian, T. Egami, K. Haule, G. Kotliar, and P. C. Dai, Phys. Rev. Lett. 112, 217202 (2014).
[36] D. Hu, X. Y. Lu, W. L. Zhang, H. Q. Luo, S. L. Li, P. P. Wang, G. F. Chen, F. Han, S. R. Banjara, A. Sapkota, A. Kreyssig, A. I. Goldman, Z. Yamani, Ch. Niedermayer, M. Skoulatos, R. Georgii, T. Keller, P. S. Wang, W. Q. Yu, and P. C. Dai, Phys. Rev. Lett. 114, 157002 (2015).
[37] J. M. Allred, K. M. Taddei, D. E. Bugaris, S. Avci, D. Y. Chung, H. Claus, C. de la Cruz, M. G. Kanatzidis, S. Rosenkranz, R. Osborn, and O. Chmaissem, Phys. Rev. B 90, 104513 (2014).
[38] M. Rotter, C. Hieke, and D. Johrendt, Phys. Rev. B 82, 014513 (2010).
[39] M. Ishikado, Y. Nagai, K. Kodama, R. Kajimoto, M. Nakamura, Y. Inamura, S. Wakimoto, H. Nakamura, M. Machida, K. Suzuki, H. Usui, K. Kuroki, A. Iyo, H. Eisaki, M. Arati, and S. I. Shamoto, Phys. Rev. B 84, 144517 (2011).
[40] C. H. Lee, P. Steffens, N. Qureshi, M. Nakajima, K. Kihou, A. Iyo, H. Eisaki, and M. Braden, Phys. Rev. Lett. 111, 167002 (2013).
[41] M. D. Lumsden, A. D. Christianson, D. Parshall, M. B. Stone, S. E. Nagler, G. J. MacDougall, H. A. Mook, K. Lokshin, T. Egami, D. L. Abernathy, E. A. Goremychkin, R. Osborn, M. A. McGuire, A. S. Sefat, R. Jin, B. C. Sales, and D. Mandrus, Phys. Rev. Lett. 102, 107005 (2009).
[42] S. Chi, A. Schneidewind, J. Zhao, L. W. Harriger, L. Li, Y. Luo, G. Cao, Z. Xu, M. Loewenhaupt, J. Hu, and P. C. Dai, Phys. Rev. Lett. 102, 107006 (2009).
[43] D. S. Inosov, J. T. Park, P. Bourges, D. L. Sun, Y. Sidis, A. Schneidewind, K. Hradil, D. Haug, C. T. Lin, B. Keimer, and V. Hinkov, Nat. Phys. 6, 178 (2010).
[44] H. Q. Luo, Z. Yamani, Y. C. Chen, X. Y. Lu, M. Wang, S. L. Li, T. A. Maier, S. Danilkin, D. T. Adroja, and P. C. Dai, Phys. Rev. B 86, 024508 (2012).
[45] Y. Zhang, Z. R. Ye, Q. Q. Ge, F. Chen, Juan Jiang, M. Xu, B. P. Xie, and D. L. Feng, Nat. Phys. 8, 371 (2012).
[46] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.94.094504 for additional data and analysis.
[47] L. W. Harriger, M. S. Liu, H. Q. Luo, R. A. Ewings, C. D. Frost, T. G. Perring, and P. C. Dai, Phys. Rev. B 86, 144043(R) (2012).
[48] Yu Li, Z. P. Yin, X. C. Wang, D. W. Tam, D. L. Abernathy, A. Podlesnyak, C. L. Zhang, M. Wang, L. Y. Xing, C. Q. Jin, K. Haule, G. Kotliar, T. A. Maier, and P. C. Dai, Phys. Rev. Lett. 116, 247001 (2016).
[49] Z. Diao, D. Campanini, L. Fang, W.-K. Kwok, U. Welp, and A. Rydh, Phys. Rev. B 93, 014509 (2016).