Iron-based superconductors (FeSCs) represent the second class of high-$T_c$ materials after the first discovery of high-$T_c$ superconductivity in cuprate materials. Both families have similar phase diagrams, in which superconductivity emerges in the vicinity of an antiferromagnetically (AFM) ordered phase. This has led to the suggestion of a spin-fluctuation mediated pairing mechanism, which is currently considered as a common thread for unconventional superconductivity.\(^1\)

By contrast, a comparison between the two classes of high-$T_c$ families shows important differences as well. Most importantly, unlike the cuprates, the parent compounds of FeSCs are metals with a spin-density-wave (SDW) ground state, which, analogously to the SDW state in Cr metal, invokes a Fermi surface (FS) nesting picture for the origin of the magnetism. Indeed, angle-resolved photoemission spectroscopy (ARPES) showed that all FeSCs share a similar band structure characterized by the presence of quasi-nested FSs,\(^2\) which enhances the tendency toward stripe-type AFM instability. The resulting AFM order has also been confirmed by neutron scattering.\(^6\)\(^-\)\(^10\) These experimental results, as well as the superconducting gap symmetry and structure of FeSCs, can be well explained within the framework of unconventional superconductivity caused by spin fluctuations of itinerant electrons.\(^11\)\(^-\)\(^13\) One can thus expect that FeSCs fall in the category of itinerant electron systems, in contrast to the case of cuprate superconductors, in which Mott physics is more fundamentally tied to superconductivity.

However, while the view based on itinerant electrons is quite successful in FeSCs, there are important reasons to expect that strong correlation physics may play an important role. First, the reduced Drude spectral weight in optical conductivity together with bad metallic behavior indicates the strongly correlated nature of FeSCs.\(^14\)\(^,\)^\(^15\) Secondly, density functional theory (DFT) calculations often fail to correctly describe the ARPES spectra of these materials owing to strong renormalization of the bands around the Fermi level.\(^16\)\(^-\)\(^19\) Both features are hallmarks of correlated metals in close proximity to a Mott insulating phase. In view of these facts, one may now pose the following important question: To what extent are FeSCs strongly correlated? Therefore, it is of particular interest to characterize the strength of the electron correlations that influence the underlying electronic and magnetic structures of FeSCs.

Herein, we report an inelastic neutron scattering (INS) study on single crystals of hole-doped Ba$_{0.75}$K$_{0.25}$Fe$_2$As$_2$ that characterizes the strength of electron correlations in FeSCs from the viewpoint of spin dynamics. By combining ARPES measurements and first-principles calculations, we show that the measured spin excitation energies are reduced by a factor of $\sim 3$ compared to those obtained from the DFT-derived model. The observation of the effective (i.e., renormalized by electron correlations) spin excitation can easily be understood as an extension of the concept of mass renormalization to dynamical spin susceptibility. Our results reveal the strongly correlated nature of FeSCs beyond the DFT level, which must be considered for realistic treatment of the spin dynamics in these materials.

Single crystals of Ba$_{0.75}$K$_{0.25}$Fe$_2$As$_2$ were grown using the FeAs-flux method, as described elsewhere.\(^20\) To avoid a reaction with vaporized potassium, the starting materials, Ba, K, and FeAs, which were placed in an aluminum crucible, were sealed in a stainless steel container.\(^21\) Transport measurements show the AFM transition at $T_N = 63$ K, followed by the superconducting transition at $T_c = 28$ K. We co-aligned 4.0 g of single crys-
Taking 27 by using incident The measurements at 4SEASONS were and placed In some cases, they The resulting INS with corresponding energy resolutions at the elastic line. The integration ranges of $L$ at $Q = (1, 0, L)$ are shown for each constant-energy map, respectively. The $|Q|$-dependent radially symmetric background was subtracted from the raw spectra. For $E > 100$ meV, data from symmetry equivalent positions in reciprocal space are averaged to improve statistics and folded into the area marked by the red dashed box. Constant-energy maps in (d)-(h) are obtained by symmetrizing the INS data in the red box. (i) Constant-energy cuts of the spin excitations along the $(1, K)$ direction at the indicated energy transfers. The black solid lines represent Gaussian fits to the data.

tals of Ba$_{0.75}$K$_{0.25}$Fe$_2$As$_2$ with a mosaic spread of 5°. INS measurements were performed using the 4SEASONS time-of-flight (TOF) chopper spectrometer at Japan Proton Accelerator Research Complex (J-PARC). Taking advantage of the repetition rate multiplication (RRM) technique for pulsed neutron sources, a set of incident neutron energies ($E_i$’s) can be obtained in one experimental run, which allows for the simultaneous measurement of the low and high energy features of an excitation spectrum. The measurements at 4SEASONS were performed above $T_c$ at $T = 30$ K by using incident neutron energies of $E_i = 300, 80.7, 36.8,$ and 13.5 meV, with corresponding energy resolutions at the elastic line of $\Delta E = 40, 6.5, 2.3$ and 0.7 meV, respectively (full-width at half-maximum). The INS data were collected over a period of 3 days using a fixed sample geometry with the $c$-axis parallel to the incident neutron beam. Data reduction of the neutron event data was performed using the UTSUSEMI software package. The resulting INS data were corrected for $|Q|$-dependent radially symmetric background from the sample environment and placed on an absolute intensity scale (mbarn sr$^{-1}$meV$^{-1}$f.u.$^{-1}$) by using a vanadium standard. In some cases, they were smoothed by convolution with a Gaussian kernel. Throughout this paper, we define the momentum transfer $Q = Ha^* + Kb^* + Le^* = (H, K, L)$ in reciprocal lattice units (r.l.u.) by using the orthorhombic unit cell. In this notation, low-energy spin excitations associated with the stripe-type AFM order occur at the in-plane wave vectors of $Q_{\text{AFM}} = (\pm 1, 0)$ and $(0, \pm 1)$. ARPES experiments were performed at BL5U of the UVSOR-III Synchrotron by using tunable linearly polarized light of $h\nu = 60$ eV. Clean sample surfaces were obtained for the ARPES measurements by cleaving single crystals in-situ in an ultrahigh vacuum better than $1 \times 10^{-8}$ Pa. The measurements were performed at $T = 6$ K.

Figures 1(a)-(h) compare the two-dimensional constant energy maps of spin excitations in the $(H, K)$ scattering plane for various energy transfers. At low energies below 60 meV [Figs. 1(a)-(c)], spin excitations peak strongly at $Q_{\text{AFM}}$, which corresponds to the nesting vector between hole and electron FSs. As the energy increases, spin excitations form transversely elongated ellipses that lead to splitting into two branches [Figs. 1(d) and (e)]. At even higher energies above 150 meV, these excitations broaden rapidly and form broad circular shapes centered at the zone boundary $Q = (\pm 1, \pm 1)$ [Figs. 1(f)-(h)]. These dispersive features are also confirmed by the constant energy cuts along the $(1, 0) \rightarrow (1, \pm 1)$ symmetry direction as shown in Fig. 1(i), where a single commensurate peak centered at $(1, 0)$ at low energies ($E < 60$ meV) splits into a pair of two peaks with increasing energy, and eventually moves close to the zone boundary $Q = (1, 1)$ at $E \sim 200$ meV.

The dispersive spin-wave-like structure can be seen more clearly in the $Q$-$E$ maps. Figures 2(a) and 2(b)
compare the low- and high-energy features of spin excitations projected along the \((1, K)\) high-symmetry direction. The spin excitations at low energies are seen to be steeply dispersing and concentrated solely in the region near \(Q_{\text{AFM}}\) [Fig. 2(a)]. As the energy increases, they disperse along the \((1, 0) \rightarrow (1, \pm 1)\) symmetry direction until the energy reaches \(E \sim 200\) meV near the zone boundary [Fig. 2(b)]. The observed spin excitation bandwidth of \(\text{Ba}_{0.75}\text{K}_{0.25}\text{Fe}_2\text{As}_2\) is similar to that reported for the parent and electron-doped \(\text{BaFe}_2\text{As}_2\) systems.\(^{28-36}\)

To understand the INS data, we provide a first-principles analysis of the spin excitation spectrum of FeSCs on the basis of the itinerant picture. As the first step, we obtain the DFT band structure of \(\text{BaFe}_2\text{As}_2\) by using the QUANTUM ESPRESSO package\(^{37}\) with the experimental lattice parameters.\(^{38}\) Here, we adopt the generalized gradient approximation (GGA) exchange-correlation functional,\(^{39}\) and take cutoff energy of \(E_{\text{cut}} = 40\) Ry and 512 \(k\)-point mesh. Then, we construct an effective five-orbital tight-binding model by using the maximally localized Wannier functions (MLWFs)\(^{40-42}\) and the unfolding procedure developed in Ref.[43]. The refolded band structure of the five-orbital model shows good agreement with the result of the original DFT calculations [Fig. 3(a)]. The effect of K substitution in \(\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2\) is treated by the rigid-band shift of the Fermi level, as its validity has been confirmed by the ARPES study.\(^{44}\) Considering Hubbard-type interactions (i.e., the intraorbital Coulomb repulsion \(U\), interorbital Coulomb repulsion \(U’\), Hund’s coupling \(J\) and pair hopping \(J’\)), we obtain the dynamical spin susceptibility \(\chi_\omega(q, \omega)\) within the random phase approximation (RPA) as,

\[
\chi_\omega(q, \omega) = \chi_0(q, \omega)[I - S\chi_0(q, \omega)]^{-1}.
\]

Here, \(S\) is the corresponding interaction vertex matrix\(^{45}\) and \(\chi_0(q, \omega)\) is the irreducible susceptibility given as

\[
\chi_0^{m_1m_2l_1l_2}(q, \omega) = \sum_{k, n, m} \frac{f(\epsilon_k^{n} + q) - f(\epsilon_k^{m})}{\omega + i\delta - \epsilon_k^{m} + \epsilon_k^{m} - \epsilon_k^{n}} \times U_{1,n}(k + q) U_{1,m}(k) U_{m,l_2}^{\dagger}(k) U_{n,l_1}^{\dagger}(k + q),
\]

where \(f(\epsilon), \epsilon_k^{m}\) and \(U_{1,n}(k)\) are, respectively, the Fermi distribution function, energy dispersion, and elements of the unitary matrices from the orbital to the band basis. For the results shown below, we set \(U = 1.0\) eV, \(U’ = U - 2J, J = J’ = U/8, T = 1.5 \times 10^{-2}\) eV, \(256 \times 256 \times 1\) \(k\)-point meshes, and smearing factor of \(\delta = 1.6 \times 10^{-2}\) eV.

Figure 3(b) shows a contour plot of the calculated \(\chi_\omega(q, \omega)\) along the high-symmetry directions. The spin excitations are markedly different in the two directions \((1, 0) \rightarrow (1, 1)\) and \((1, 0) \rightarrow (0, 0)\), which is consistent with the transversely elongated spin excitations seen in Fig. 1. Such highly anisotropic spin excitations are typical of FeSCs.\(^{28-34,36,46,47}\) However, while our RPA calculation reproduces the dispersive spin-wave-like feature along the high-symmetry directions, it apparently overestimates the energy scale of the excitations.

Along the \((1, 0) \rightarrow (1, 1)\) direction, the theoretical spin excitation peak extends nearly to \(E \sim 600\) meV, which is larger by a factor of \(\sim 3\) than the experimental data.

This overestimation of the spin excitation energy in our RPA analysis has important implications for the electronic state of FeSCs. As can be seen in Eq. (2), the electronic band structure \(\epsilon_k^{m}\) determines the momentum- and energy-dependent structure of the dynamical spin susceptibility. The discrepancy between the experimental and theoretical spin excitations, therefore, suggests that the actual electronic structure of \(\text{Ba}_{0.75}\text{K}_{0.25}\text{Fe}_2\text{As}_2\) deviates from the DFT-derived model. In principle, DFT provides a good starting point for modeling the electronic structure of the weakly correlated regime. However, when electron correlations become sizable, the low-energy bands near the Fermi level are heavily renormalized, which results in a substantial effective mass \((m^*)\) enhancement (or equivalently, bandwidth \((W)\) narrowing) relative to DFT calculations. Spectroscopic probes such as ARPES can provide direct information about the real single-particle spectra of the correlated materials, which cannot be accurately captured by DFT.

To gain more insight into the experimental electronic structure, we performed ARPES measurements on crystals from the same batch as that used for INS measurements. As expected for the correlated state, mass renormalization relative to the DFT calculations \((m^*/m_{\text{DFT}})\), which quantifies the strength of electron correlation, was clearly observed. Figure 3(c) shows the spectral image of \(\text{Ba}_{0.75}\text{K}_{0.25}\text{Fe}_2\text{As}_2\) along the \(Z\)-\(X\) high-symmetry direction overlaid with DFT bands. The high-intensity region at the \(X\)-point corresponds to the bottom of the \(d_{xz/yz}\) electron band. The overall
This analysis interestingly the obtained mass enhancement and $T_l$ which is comparable and in the future INS Our results, taken together with $m^{\ast}$ that the $Fe-3d$ bandwidth narrowing is estimated by scaling the DFT band to fit the $d_{xz/yz}$ band bottom at X. This analysis yields $m^{\ast}/m_{DFT} \sim 3$, which is in reasonable agreement with estimates from the fluctuation exchange (FLEX) approximation. Interestingly, the obtained mass enhancement of $m^{\ast}/m_{DFT} \sim 3$ is surprisingly close to the renormalization factor of the spin excitation bandwidth. Such consistency between the results of INS and ARPES has not been reported before. Given the strong sensitivity of the spin excitations to the underlying electronic structure, one can expect that the $Fe-3d$ bandwidth narrowing due to electron correlations is directly reflected in the spin excitation energy scale. To confirm this, we reevaluated the dynamical spin susceptibility under the scaling of the electron band energy as $\varepsilon_k^{m} \rightarrow \varepsilon_k^{m}/z$. (Here, $z = m^{\ast}/m_{DFT}$ is the ARPES-derived mass enhancement factor.) In addition, we applied similar renormalization to the on-site interactions, temperature, and smearing factor. As shown in Fig. 3(d), this scaling reduced the spin excitation bandwidth to $\sim 1/3$ of its original width, yielding a broadly consistent description of the observed INS data. The reasons for this consistency can easily be understood as follows. The dispersion of spin excitations is defined by the resonance condition in Eq. (2), in which the denominator of irreducible susceptibility becomes zero. If the electron band energy is renormalized as $\varepsilon_k^{m} \rightarrow \varepsilon_k^{m}/z$, correspondingly, a spin excitation peak energy, defined as $\varepsilon_{k+q}^{n} - \varepsilon_{k}^{m}$, shows similar renormalization, much like the electron bands. This means that the concept of mass renormalization in the Fermi-liquid theory can be extended to dynamical spin susceptibility. A similar discussion can be used to understand the material- and doping-dependent trends of spin-excitation bandwidth in FeSCs, as has been shown using the dynamical mean field theory (DMFT).

Our results thus demonstrate that it is possible to model the spin excitations of FeSCs by incorporating aspects of the low-energy quasiparticle renormalization that affect both single- and two-particle quantities. In addition, the consistency of the mass renormalization factors determined by independent INS and ARPES measurements highlights the potential capability of INS for characterizing the strength of electron correlations. The observed mass renormalization $m^{\ast}/m_{DFT} \sim 3$ is comparable to that of typical correlated metals such as SrVO$_3$ ($m^{\ast}/m_{DFT} \sim 2$) and Ti$_2$Ba$_2$CuO$_{6+\delta}$ ($m^{\ast}/m_{DFT} \sim 3$). In this context, it is interesting to recall a recent first-principles study that estimated the strength of electron correlations, $U/t$ ($U/W$), by using the constrained RPA method. (Here, $t$ is the nearest-neighbor hopping parameter.) This calculation yields $U/t$ ($U/W$) = 8–14 (0.5–0.9) for FeSCs as an average of the five orbitals which is comparable to or even larger than $U/t$ ($U/W$) = 2–7 (0.2–0.8) obtained for cuprates. Our results, taken together with these considerations, suggest that FeSCs have stronger electron correlations than previously expected and more importantly, such a correlated electronic state is a crucial aspect that must be considered to realistically describe spin dynamics. With recent advances in modern INS spectrometers at spallation neutron sources, it is now becoming possible to experimentally determine the complicated spin susceptibility arising from the correlated band structure, and in the future INS will allow us to discuss both magnetic and electronic structures on an equal footing.

To summarize, we performed a combined INS and ARPES study on Ba$_{0.75}$K$_{0.25}$Fe$_2$As$_2$ that revealed the effects of electron correlations on spin dynamics in
FeSCs. The measurements show, in combination with first-principles calculations, that the correlation-induced narrowing of the Fe-3d bandwidth is reflected directly in the spin-excitation bandwidth. Our analysis of the spin excitation spectrum provides much richer information on the nature of electron correlations than can be obtained in a conventional analysis based on the spin-only Hamiltonian. In addition, the two independent momentum-resolved techniques used in the present study, INS and ARPES, are closely related, and provide the same mass renormalization factors consistently. These results highlight the potential of INS for use as a momentum-resolved probe for determining the electronic structure of correlated electron systems.

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However, one should note that, in the renormalization.

A simple rescaling of the DFT bands is not enough to fully account for the ARPES data. We therefore used the energy position of the band bottom at the X point as an overall measure of the band renormalization. We note that while the ARPES measurements were performed below $T_c$, the overall electron bandwidth is not affected by the occurrence of superconductivity.

Here, we mention that the mass-renormalized spin susceptibility exhibits a peak structure at around $E \sim 500$ meV, which is higher than the experimentally accessible energy range of the present study. This excitation is consistent with a previous study of the 1111 system without mass renormalization. However, one should note that, in the RPA treatment, the dynamical effects of the quasiparticle lifetime are neglected. Therefore, the experimental spin excitations at such high energies would largely be obscured by the finite quasiparticle lifetime.