Competing Magnetic Fluctuations in Iron Pnictide Superconductors:
Role of Ferromagnetic Spin Correlations Revealed by NMR

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In the iron pnictide superconductors, theoretical calculations have consistently shown enhancements of the static magnetic susceptibility at both the stripe-type antiferromagnetic and in-plane ferromagnetic (FM) wave vectors. However, the possible existence of FM fluctuations has not yet been examined from a microscopic point of view. Here, using ^{75}As NMR data, we provide clear evidence for the existence of FM spin correlations in both the hole- and electron-doped $BaFe_2As_2$ families of iron-pnictide superconductors. These FM fluctuations appear to compete with superconductivity and are thus a crucial ingredient to understanding the variability of T_c and the shape of the superconducting dome in these and other iron-pnictide families.

The role of magnetic fluctuations in iron pnictide superconductors (SCs) has been extensively studied since their discovery. As the parent materials have antiferromagnetic (AFM) ground states, attention has been understandably focused on stripe-type AFM fluctuations, which are widely believed to give rise to the Cooper pairing in these systems. In the standard picture, carrier doping or pressure application results in suppression of the AFM order and the emergence of a SC state, with T_c ranging from a few K to 56 K [1]. However, as of yet, there is no accepted theory for T_c in these materials with which to explain the large variability in maximum T_c between different iron arsenide families and the different shapes of the SC dome with electron and hole doping.

Recent nuclear magnetic resonance (NMR) measurements on non-SC, paramagnetic (PM) $SrCo_2As_2$, the $x = 1$ member of the electron-doped $Sr(Fe_{1-x}Co_x)_2As_2$ family, revealed strong ferromagnetic (FM) spin fluctuations in the Co layer coexisting with stripe-type AFM fluctuations [2,3]. Since stripe-type AFM fluctuations are a key ingredient to SC in the iron pnictides, this result suggested that FM fluctuations might compete with the stripe-type AFM fluctuations, suppressing SC in $SrCo_2As_2$. FM correlations were also observed in isostructural $BaCo_2As_2$ [2,4]. Similarly, $CaCo_{1.86}As_2$ has an A-type AFM ground state with in-plane FM order [5]. These results also raise the question of whether similar FM correlations exist generally in the SC $A(Fe_{1-x}Co_x)_2As_2$ compounds, not just at the $x = 1$ edges of their phase diagrams.

According to density functional theory calculations [6–10], the generalized static magnetic susceptibility $\chi(\mathbf{q})$ is enhanced at both the FM and stripe-type AFM wave vectors in all the iron-based SCs and parent compounds. Experimentally, the uniform $\chi(\mathbf{q} = 0)$ of the parent compounds is enhanced by a factor of order 5 over band structure values, which is consistent with FM

correlations [1]. Nevertheless, FM fluctuations have not been investigated microscopically, perhaps because low-energy FM fluctuations are difficult to observe via inelastic neutron scattering (INS). The peak in the inelastic structure factor at $\mathbf{q} = 0$ coincides with the elastic Bragg diffraction peaks, and the energy scale of thermal neutrons is relatively high. The study of low-energy FM fluctuations therefore requires cold, polarized neutrons. NMR, in contrast, is a microscopic probe uniquely sensitive to low-energy FM fluctuations via the modified Korringa ratio.

In this Letter, using ^{75}As NMR measurements, we present clear evidence for FM fluctuations in the tetragonal, PM phase of both the hole- and electron-doped $BaFe_2As_2$ families of iron pnictide SCs. Furthermore, we suggest that these FM fluctuations compete with SC, and that this competition between FM and AFM fluctuations may be a key ingredient to a theory of T_c in the iron pnictides.

For this study, we chose $x = 4.7\%$ ($T_N \sim 50$ K and $T_c \sim 15$ K) and $x = 5.4\%$ ($T_N \sim 35$ K and $T_c \sim 20$ K) in single-crystalline $Ba(Fe_{1-x}Co_x)_2As_2$ as representative superconducting samples in which to look for FM correlations. We also used our existing data on $BaCo_2As_2$, reported elsewhere [2,11] and other data from the literature. The ^{75}As NMR shift and spin-lattice relaxation rates $1/T_1$ were measured under magnetic fields parallel to the c axis ($H \| c$) and to the ab plane ($H \| ab$).

Figures 1(a) and 1(b) show the existing NMR data (T dependence of NMR shift K and $1/T_1T$, respectively) for both the electron-doped $Ba(Fe_{1-x}Co_x)_2As_2$ and hole-doped $Ba_{1-x}K_xFe_2As_2$ families of iron-pnictide SCs. The NMR shift consists of a T-independent orbital shift K_0 and a T-dependent spin shift $K_{spin}(T)$ due to the uniform magnetic spin susceptibility $\chi(\mathbf{q} = 0)$. The NMR shift can therefore be expressed as $K(T) = K_0 + K_{spin}(T) = K_0 + A_{hf}\chi_{spin}/N_A$, where N_A is Avogadro's number, and A_{hf} is the hyperfine coupling, usually expressed in units of
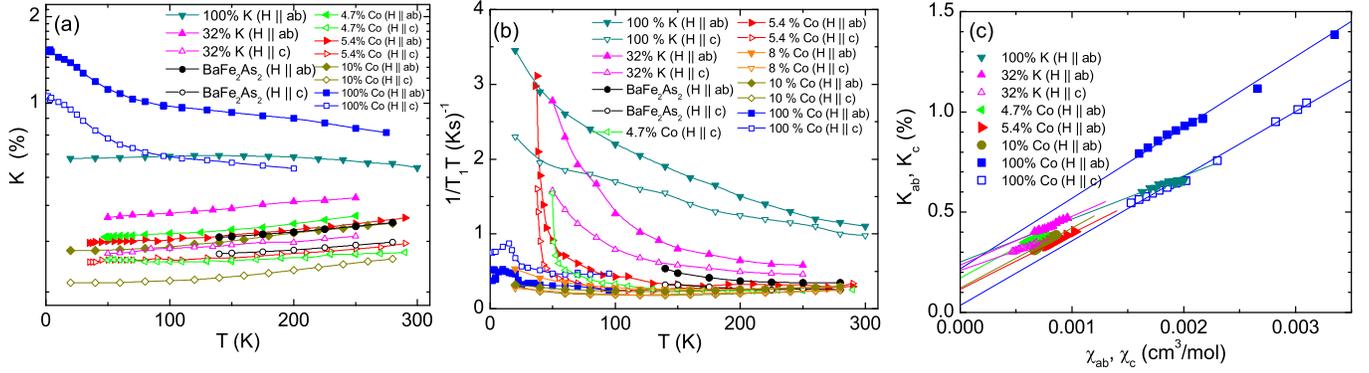
FIG. 1 (color online).   (a) T dependence of the NMR shift K for a variety of indicated samples. (b) T dependence of NMR spin-lattice relaxation rate $1/T_1 T$ for the same samples. Here, and throughout, filled (open) symbols are used for $H \| ab$ ($H \| c$). (c) $K_i(T)$ vs $\chi_i(T)$ ($i = ab, c$) for a variety of samples. Data are from Refs. [14,19,23,34–38]. For KFe$_2$As$_2$, K_c is nearly T independent making a K vs χ analysis impossible. From the relative values of K_{ab} and K_c, we estimate $K_{0,c} \sim 0.21\%$. For BaFe$_2$As$_2$, $K_{0,ab} = 0.14\%$ and $K_{0,c} = 0.21\%$ are from Ref. [14]. For the 8% Co doped sample, $K_{0,c} = 0.22\%$ from Ref. [19].

kOe/μ_B. In order to extract $K_{\text{spin}}(T)$, we plot $K(T)$ against the corresponding bulk static uniform magnetic susceptibility $\chi(T)$ with T as shown in Fig. 1(c). From the y intercept of the linear fit curve, we can estimate the orbital shift K_0, and extract $K_{\text{spin}}(T)$ needed for the following Korringa ratio analysis.

To extract the character of spin fluctuations in the iron pnictides from ^{75}As NMR data, we employ a modified Korringa ratio analysis. Within a Fermi liquid picture, both $1/T_1 T$ and K_{spin} are determined primarily by the density of states at the Fermi energy $\mathcal{D}(E_F)$, leading to the Korringa relation $T_1 T K_{\text{spin}}^2 = (\hbar/4\pi k_B)(\gamma_e/\gamma_N)^2 \equiv S$. For the ^{75}As nucleus ($\gamma_N/2\pi = 7.2919$ MHz/T), $S = 8.97 \times 10^{-6}$ Ks. Deviations from $T_1 T K_{\text{spin}}^2 = S$, which are conveniently expressed via the Korringa ratio $\alpha \equiv S/(T_1 T K_{\text{spin}}^2)$, can reveal information about electron correlations in the material [12,13]. For uncorrelated electrons, we have $\alpha \sim 1$. However, enhancement of $\chi(\mathbf{q} \neq 0)$ increases $1/T_1 T$ but has little or no effect on K_{spin}, which probes only the uniform $\chi(\mathbf{q} = 0)$. Thus $\alpha > 1$ for AFM correlations. In contrast, $\alpha < 1$ for FM correlations. The Korringa ratio α, then, reveals whether the magnetic correlations in the material have predominantly FM or AFM character.

To perform the Korringa ratio analysis, one needs to take the anisotropy of K_{spin} and $1/T_1 T$ into consideration. The $1/T_1$ probes hyperfine field fluctuations at the NMR Larmor frequency, ω_N, perpendicular to the external magnetic field according to $(1/T_1)_{H\|i} = \gamma_N^2[|H_j^{\text{hf}}(\omega_N)|^2 + |H_k^{\text{hf}}(\omega_N)|^2]$, where (i, j, k) are mutually orthogonal directions and $|H_j^{\text{hf}}(\omega)|^2$ represents the power spectral density of the jth component of the hyperfine magnetic field at the nuclear site. Thus, defining $H_{ab}^{\text{hf}} \equiv H_a^{\text{hf}} = H_b^{\text{hf}}$, which is appropriate for the tetragonal PM state, we have $(1/T_1)_{H\|c} = 2\gamma_N^2|H_{ab}^{\text{hf}}(\omega_N)|^2 \equiv 1/T_{1,\perp}$. The Korringa parameter $\alpha_\perp \equiv S/T_{1,\perp} T K_{\text{spin},ab}^2$ will then characterize

fluctuations in the ab-plane component of the hyperfine field. By analogy, we should pair $K_{\text{spin},c}$ with $2\gamma_N^2|H_c^{\text{hf}}(\omega_N)|^2 \equiv 1/T_{1,\|}$, so that the Korringa parameter $\alpha_\| = S/T_{1,\|} T K_{\text{spin},c}^2$ characterizes fluctuations in the c-axis component of the hyperfine field. Since $(1/T_1)_{H\|ab} = \gamma_N^2[|H_{ab}^{\text{hf}}(\omega_N)|^2 + |H_c^{\text{hf}}(\omega_N)|^2]$, we estimate the quantity $1/T_{1,\|} T$ from $1/T_{1,\|} T = 2(1/T_1 T)_{H\|ab} - (1/T_1 T)_{H\|c}$.

The T dependences of the Korringa ratios $\alpha_\perp = S/T_{1,\perp} T K_{\text{spin},ab}^2$ and $\alpha_\| = S/T_{1,\|} T K_{\text{spin},c}^2$ are shown in Fig. 2(a). In BaCo$_2$As$_2$, both $\alpha_\perp$ and $\alpha_\|$ are nearly independent of T and much less than 1, consistent with FM correlations. For the remaining samples, $\alpha_\|$ is generally greater than 1 indicating AFM correlations throughout the
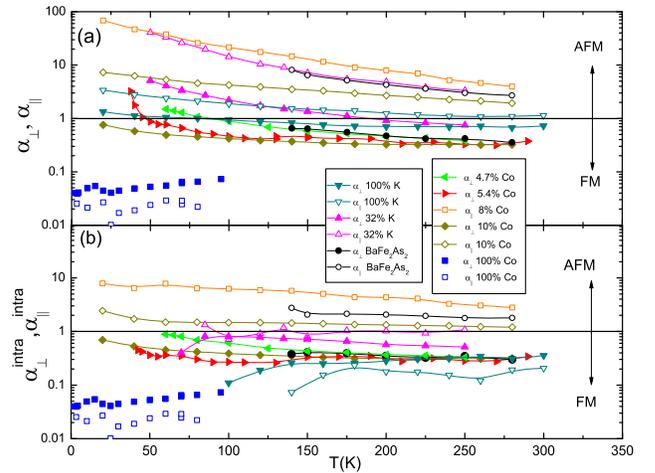

FIG. 2 (color online).   (a) Korringa ratios $\alpha_\perp$ (filled symbols) and $\alpha_\|$ (open symbols) as a function of T in a variety of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ and Ba$_{1-x}$K$_x$Fe$_2$As$_2$ samples. (b) Same for intraband Korringa ratios $\alpha_\|^{\text{intra}}$ and $\alpha_\perp^{\text{intra}}$, obtained by subtracting the interband (Curie-Weiss) contributions.

T range. In addition, both $\alpha_\perp$ and $\alpha_\parallel$ increase as T is lowered, showing the growth of AFM spin fluctuations at low T. In contrast, we find that $\alpha_\perp \sim 0.3 < 1$ for the parent and Co-doped samples in the high-T PM phase. The hole-doped $Ba_{1-x}K_xFe_2As_2$ also display $\alpha_\perp \lesssim 1$ in the PM phase, suggesting FM correlations, although less strong than in the Co-doped samples.

Careful consideration is required to interpret the value of the Korringa ratio. In comparing the α value to the cross-over $\alpha_0 = 1$ between dominant FM and AFM fluctuations, one is assuming a simple model in which the nuclear relaxation is due to the local $\mathcal{D}(E_F)$ at the As sites through on-site hyperfine interactions, where As-$4p$ bands hybridize with Fe-$3d$ bands [14]. If, on the other hand, the nuclear relaxation is induced only by the localized Fe spins through isotropic transferred hyperfine interactions, the value of α would instead be compared to the crossover $\alpha_0 = 1/4$, assuming no contributions to $1/T_1$ from AFM correlations due to form factor effects [15–17]. In the highly overdoped $x = 26\%$ $Ba(Fe_{1-x}Co_x)_2As_2$, however, AFM fluctuations are known to be absent from INS measurements [18]. Accordingly, Refs. [1] and [19] find $\alpha \sim 1.2$, suggesting weak correlation. If the crossover were $\alpha_0 = 1/4$, this value of 1.2 for the Korringa ratio must be associated with dominant AFM fluctuations, in conflict with observations. These results suggest that the factor of 4 change to $\alpha_0 = 1/4$ proposed by Ref. [16] for iron pnictides is too large. In fact, the FM correlations have been also pointed out in $(La_{0.87}Ca_{0.13})FePO$ with $\alpha = 0.37$ by ^{31}P NMR [20]. In addition, in the case of Na_xCoO_2 for $x > 0.65$, FM correlations are known to be present [17] and the measured Korringa ratio takes the value $\alpha \sim 0.3$ [21]. It is also noted that the Wilson ratio for $BaFe_2As_2$ is mildly enhanced ($R_W \sim 3$) [22], consistent with FM correlations. Thus we conclude that the value we observe, $\alpha_\perp \sim 0.3$, can be reasonably attributed to FM fluctuations.

To discuss the magnetic correlations based on the values of $\alpha_\perp$ and $\alpha_\parallel$ in the iron pnictides in more detail, it is helpful to consider the hyperfine field at the ^{75}As site, which is determined by the spin moments on the Fe sites through the hyperfine coupling tensor [23]. In this case, there are two sources of hyperfine field pointing along the c axis [14]: stripe-type AFM fluctuations at $q = (\pi, 0)/(0, \pi)$ with the spins pointing within the ab plane [as illustrated in Fig. 3(a)] or FM fluctuations at $q = 0$ with the spins

pointing along the c axis [Fig. 3(b)]. Similarly, hyperfine field fluctuations in the ab plane can result from FM fluctuations at $q = 0$ with the spins pointing within the ab plane [Fig. 3(c)], or from AFM fluctuations at $q = (\pi, 0)/(0, \pi)$ with the spins pointing along the c axis [Fig. 3(d)]. Thus, the value of $\alpha_\parallel$ reflects the competition between (a)- and (b)-type correlations: type (a) AFM correlations will increase $\alpha_\parallel$ above 1, while type (b) FM correlations will lower $\alpha_\parallel$ below 1. Similarly, $\alpha_\perp$ reflects the competition between (c)- and (d)-type correlations: type (d) AFM correlations will increase $\alpha_\perp$, while type (c) FM correlations will lower $\alpha_\perp$.

In what follows, we will refer to the correlations depicted in Fig. 3(a) as "type (a)" correlations (similarly for the others). Since $\alpha_\parallel$ reflects the character of the c-axis component hyperfine field fluctuations, the AFM value of $\alpha_\parallel$ in Fig. 2 can be attributed to type (a) correlations, i.e., stripe-type AFM correlations with the Fe spins in-plane. These must dominate type (b) fluctuations in order to have an AFM value of $\alpha_\parallel$. Similarly, since $\alpha_\perp$ reflects the character of the ab-plane component of hyperfine field fluctuations, the FM value of $\alpha_\perp$ in the high-T region can be attributed to type (c) in-plane FM fluctuations. On the other hand, the increase of $\alpha_\perp$ as the temperature is lowered reflects the increasing dominance of type (d) stripe-type AFM correlations with a c-axis component to the spin. This clearly indicates the simultaneous coexistence of FM and AFM fluctuations. Furthermore, the dominance of type (a) and (c) spin fluctuations in the high-T region suggests that both the AFM and FM fluctuations are highly anisotropic in the iron pnictides, favoring the ab plane.

Finally it is interesting to isolate the FM fluctuations and extract their T dependence. We adopt the simple phenomenological model of Refs. [19,24,25] to decompose $1/T_1T$ into inter- and intraband components according to $1/T_1T = (1/T_1T)_{inter} + (1/T_1T)_{intra}$. The T dependence of the interband term is assumed to follow the Curie-Weiss form appropriate for 2D AFM fluctuations: $(1/T_1T)_{inter} = C/(T - \Theta_{CW})$. For the Co-doped samples, we use $(1/T_1T)_{intra} = \alpha + \beta \exp(-\Delta/k_BT)$, while for the K-doped samples we simply use $(1/T_1T)_{intra} = $ const, as in Ref. [23]. The Curie-Weiss parameter C measures the strength of AFM fluctuations, and Θ_{CW} corresponds to the distance in T from the AFM instability point. Here, we decompose the quantities $1/T_{1,\parallel}T$ and $1/T_{1,\perp}T$ into their inter- and intraband components. Our results for the CW parameters $C_\perp$, $C_\parallel$, and Θ_{CW}, shown in Fig. 4, are consistent with the results of Refs. [23] and [19]. Similar carrier doping dependence of Θ_{CW} is reported in P-doped $BaFe_2As_2$ [26] and in $LaFeAsO_{1-x}F_x$ [27]. We use the intraband components to calculate the Korringa ratios $\alpha_\parallel^{intra}$ and $\alpha_\perp^{intra}$. The results are shown in Fig. 2(b). Both $\alpha_\parallel^{intra}$ and $\alpha_\perp^{intra}$ remain roughly constant through the T range. The deviations at low T are due to imperfect
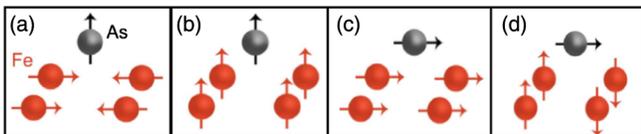

FIG. 3 (color online). (a),(b): Competing sources of hyperfine field fluctuations along the c axis. (c),(d): Competing sources of hyperfine field fluctuations in the ab plane. Competition between (a) and (b) [(c) and (d)] determines the value of $\alpha_\parallel$ ($\alpha_\perp$).
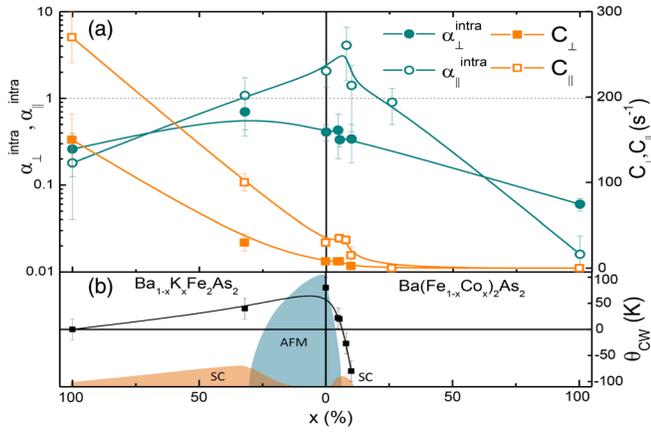
FIG. 4 (color online). Potential relevance of FM spin fluctuations to iron pnictide phase diagram: (a) Doping dependence of the nearly T-independent values of $\alpha_\parallel^{intra}$ and $\alpha_\perp^{intra}$, which parametrize the strength of FM fluctuations [39]. We also show the doping dependence of the Curie-Weiss parameters $C_\perp$ and $C_\parallel$, which parametrize the strength of 2D AFM fluctuations. (b) The doping dependence of the Curie-Weiss temperature Θ_{CW}. Solid lines are guides to the eye.

subtraction of the interband part, arising from our simplistic Curie-Weiss fitting. We notice that $\alpha_\parallel^{intra}$ for several compounds are greater than 1, suggesting AFM correlations in the intraband component. On the other hand, the value of $\alpha_\perp^{intra}$ is consistent with FM fluctuations, as discussed above, for all samples.

What then is the role of these FM fluctuations in the iron pnictide superconductors? In Fig. 4, we summarize our results across the combined hole- and electron-doped phase diagram of $BaFe_2As_2$. First of all, $C_\parallel$ is always greater than $C_\perp$ in the entire phase diagram, indicating that type (a) spin fluctuations are stronger than type (d) spin fluctuations. On the electron-doped side, AFM spin fluctuations die out beyond the SC dome at $x \sim 15\%$ [18]. In contrast, the AFM spin fluctuations become very strong on the hole-doped side relative to the electron-doped side. The doping dependences of $C_\perp$ and $C_\parallel$ are reminiscent of the doping dependence of the mass enhancement [28]. For a measure of the strength of the FM fluctuations, we plot in Fig. 4(a) the average values of $\alpha_\parallel^{intra}$ and $\alpha_\perp^{intra}$ above 150 K except for $BaCo_2As_2$ where we average over all data. We find that $\alpha_\perp^{intra}$ has a FM value throughout the phase diagram, consistent with in-plane FM [type (c)] spin fluctuations. In contrast, $\alpha_\parallel^{intra}$ shows an AFM value at 8% Co doping, but exhibits a dramatic decrease towards FM values when hole doped or electron doped beyond 8%. The FM fluctuations are thus strongest at the maximally doped edges of the phase diagram. The disappearance of AFM spin fluctuations beyond 15% Co doping coincides with the appearance of FM fluctuations, suggesting a competition between FM and AFM fluctuations. On the hole-doped

side, AFM correlations clearly increase in strength. Paradoxically, this increase in strength of AFM correlations is accompanied by a decrease of T_c, as noted in Ref. [29]. Our analysis offers a possible explanation. FM correlations also increase in strength on the hole-doped side, as seen from the rapidly decreasing values of $\alpha_\parallel^{intra}$ and $\alpha_\perp^{intra}$ and the increasing value of the NMR shift [Fig. 1(a)] with increasing hole doping. We suggest that the growth of competing FM correlations results in the reduction of T_c despite the increase in AFM correlation strength. In KFe_2As_2, then, FM and AFM correlations coexist with neither dominating the other, leading to the Korringa parameters $\alpha_\perp \sim 1$ and $\alpha_\parallel \sim 1$ that we observe in Fig. 2(a). Finally, it is noted that structural parameters have been pointed out to play an important role for controlling the ground state of iron pnictides [30]. Although we discussed our NMR data based on the well-known phase diagram where the tuning parameter is carrier doping, the observed trends should not be attributed to carrier concentration alone.

In conclusion, using an anisotropic modified Korringa ratio analysis on ^{75}As NMR data, we have provided clear evidence for the existence of FM spin correlations in both hole- and electron-doped $BaFe_2As_2$. The FM fluctuations are strongest in the maximally doped $BaCo_2As_2$ and KFe_2As_2 but are still present in the $BaFe_2As_2$ parent compound, consistent with its enhanced χ [1]. While we consider here only the Ba122 system, similar results are found for other iron-pnictide-based superconductors. In particular, FM values of α were also observed in the PM phase of $LaO_{0.9}F_{0.1}FeAs$ ($\alpha = 0.55 < 1$) [31], $K_{0.8}Fe_2Se_2$ ($\alpha = 0.45 < 1$) [32], and $Ca(Fe_{1-x}Co_x)_2As_2$ [33]. These FM fluctuations appear to compete with superconductivity and are thus a crucial ingredient for understanding the variability of T_c and the shape of the SC dome. Our results indicate that theoretical microscopic models should include FM correlations to capture the phenomenology of the iron pnictides. Polarized INS experiments examining magnetic response at the FM wave vector will be needed to further understand the interplay between FM and AFM spin correlations in the iron pnictides.

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