Magnetic fluctuations in an unconventional superconductor (U-SC) can distinguish between distinct proposals for the symmetry of the order parameter. Motivated thereby, we undertake a study of magnetic fluctuations in Iron Pnictides (FePn) tracking their evolution from the incoherent normal, pseudogapped metal, to the U-SC state. Within our proposal of extended-s-plus $s_{xy}$ in-plane gap with proximity-induced out-of-plane line nodes, (i) we describe the evolution of the spin-lattice relaxation rate, from a non-Korringa form in the normal state, to a power-law form in the U-SC state in good agreement with experiment, and (ii) we predict a sharp resonance in the U-SC state along $(\pi, \pi)$, but not along $(\pi/2, 0)$, along with modulated $c$-axis intensity in inelastic neutron scattering work as a specific and testable manifestation of our proposal.

The precise mechanism of unconventional superconductivity (U-SC) in the recently discovered Iron Pnictides (FePn) is presently a hotly debated issue. While many physical responses are reminiscent of cuprates, FePn are metals, albeit presumably proximate to a Mott insulator. Moreover, relevance of all $d$ orbitals in FePn considerably complicates determination of the pair symmetry.

The central quantity of interest is the dynamical spin susceptibility, $\chi''(q, \omega)$, has been measured for the 122 FePn, showing that the SC gap has in-plane smooth angular variation and an out-of-plane $\cos(k_z c)$ component.

Extant theoretical works have studied these issues using effective model Hamiltonians, both in the weak and strong coupling limits. In the itinerant approach, the magnetic fluctuations have been computed within HF-RPA. For $s_{\pm}$ pairing, a sharp resonance for $Q = (\pi, \pi)$ in INS is predicted below $T_c$ [12], while no such feature is arises for $s$, $ex$-$s$, and $d$ wave-pairing, or for $q \neq Q$. To get the power-law-in-$T$ behavior in NMR and $\mu$SR in the $s_{\pm}$ idea, it is necessary to consider (strong) disorder effects in a two- or four-band model. Again, the situation is controversial. For LaFePO, the penetration depth, $\lambda(T) \approx T^{1.2}$ [13], while a similar study on a wide range of samples of different FePn found a seeming universality in $\lambda(T)$; this mitigates against the disorder effects [10]. If this is true, one must consider both the NMR and INS data within a theoretical scenario with out-of-plane line nodes in the SC gap, since in-plane nodes seem to be ruled out by extant tunnelling data [17].

Recently, based on inputs from the correlated normal state electronic structure and rigorous symmetry arguments, we proposed a specific gap function with sizable in-plane angular variation (but no nodes) and inter-band proximity induced out-of-plane line nodes [18]. In contrast to the itinerant picture, our proposal is based on a strong correlation view of FePn. Here, we investigate the NMR and INS response within such a correlated approach, using the full, multiband spectral functions for all $d$ orbitals. LDA+DMFT can readily access the intermediate coupling regime relevant for FePn [19]. We show how our proposal gives a quantitative account of the NMR $T_1^{-1}$ over the whole $T$ range, and makes specific predictions with regard to the observation of the low-energy dispersive resonance in the INS intensity below $T_c$.
agreement we find below is an excellent reconciliation of ARPES and INS data [22]. This might be the case, we argue that the good quantitative agreement between LDA+DMFT and key experiments in both, the normal and U-SC states, has been shown there, lending strong support for our choice. The prescription is simple: replace the band Green functions used in weak-coupling approaches [12] by their LDA+DMFT counterparts. This ensures that the dynamical aspect of strong, local, multi-orbital (MO) correlations is included from the outset.

For a MO-system, after replacing the bare $G_{aa}(k,\omega)$ with $G_{aa}(k,\omega) \equiv G^{LDA+DMFT}(k,\omega) = [\omega - \epsilon_{ka} - \Sigma_{k}(\omega)]^{-1}$ and $F_{ab}(k,\omega) = G_{aa}(k,\omega) \frac{\Delta_{ab}(k)}{\omega + \epsilon_{ka} + \Sigma_{k}(\omega)}$ and introducing the spin operator $S_{\alpha,\mu}(q) = \frac{1}{2} \sum_{k} \epsilon_{\alpha,\mu,\sigma}(k + q) c_{\alpha,\sigma,\sigma'}^{\dagger}(k)$, with $\mu = x, y, z$, the “bare” dynamical spin susceptibility reads

$$\chi_{\alpha,\sigma,\sigma'}^{\mu}(q,\omega) = -\frac{1}{2} \sigma_{\alpha,\sigma,\sigma'}^{\mu} \sigma_{\beta,\sigma,\sigma'} \sum_{k,\omega'} [G_{aa}(k + q,\omega + \omega') - G_{ab}(k,\omega') + F_{ab}(-k - q, -\omega - \omega') G_{ba}(k,\omega')]$$

Including the ladder vertex in an infinite summation of “ladder” diagrams using RPA, the renormalized magnetic susceptibility, $\chi_{\alpha,\beta}(q,\omega) = [\chi_{\alpha,\beta}(q,\omega) - J(q)]^{-1}$, where $\chi_{\alpha,\beta}(q,\omega) = \sum_{q} \chi_{\alpha,\beta}(q,\omega)$ and $J(q) = J_{1} (\cos(q_{x}) + \cos(q_{y})) + J_{2} \cos(q_{z}) \cos(q_{y})$, with $J_{1} \simeq \frac{\epsilon_{0}^{\mu}}{\epsilon_{\sigma} - J_{1}}$ and $J_{2} \simeq \frac{\epsilon_{0}^{\mu} J_{1}}{\epsilon_{\sigma} - J_{1}}$ being the frustrated superexchange scales in FePn [2]. Using $\chi_{\alpha,\beta}(\epsilon) = C \int d f(\epsilon) [1 - f(\epsilon)] W(\epsilon)$ in the RPA series, the NMR relaxation rate, $T_{1}^{-1} = \sum_{q} \chi_{\alpha,\beta}(q,\omega)|_{\omega = 0}$, can be now expressed in terms of the full DMFT propagators. Here, $W(\epsilon) = \sum_{\alpha,\beta} \{ n_{\alpha,\beta}(\epsilon) \rho_{\alpha,\beta}(\epsilon) + \rho_{\alpha,\beta}(\epsilon) n_{\alpha,\beta}(\epsilon) \}$ and the $\rho_{\alpha,\beta}(\epsilon), n_{\alpha,\beta}(\epsilon)$ are the LDA+DMFT local spectral functions computed earlier [18]. Also, $C = 2(\frac{2}{\pi})^{2} (\frac{\gamma_{r} \gamma}{\gamma_{r} \gamma_{r} + \gamma_{i}^{2}})$. Finally, our restriction to the non-crossing diagrams in the ladder approximation for $\chi(q,\omega)$ is an approximation. It is possible that “non-crossing” diagrams need to be included in a full description. However, for the underdoped cuprates, it has been shown that a renormalized “RPA” summation for $\chi(q,\omega)$ with fully renormalized one-particle $G_{\sigma}(k,\omega), F_{\sigma,\sigma'}(k,\omega)$ gives excellent reconciliation of ARPES and INS data [22]. This suggests small vertex corrections: while we cannot prove why this should be the case, we argue that the good agreement we find below is an a posteriori justification for neglecting them in our theory.

The NMR spin-lattice relaxation rate is a measure of the local spin fluctuation rate in both phases. For an s-wave SC, the coherence factors give the Hebel-Slichter (HS) enhancement as a peak in $T_{1}^{-1}$ below $T_{c}$. When the “normal” state is strongly incoherent (large Im$\Sigma(\omega = E_{F}) \neq 0$, as in our case), or the SC gap has nodes [3], the HS peak is absent. But $T_{1}^{-1} \approx e^{-\Delta_{c} EF}$ survives for $T < c$ in an s-wave SC, while a power-law fall-off in $T$ characterizes an U-SC with gap nodes [3,4]. In the normal state above $T_{c}$, we set $F_{ab}(k,\omega) = 0$. This suffices for computing the NMR $T_{1}^{-1}$. More work has to be done to compute the INS intensity; we will present details in a separate work.

However, qualitative remarks about what we expect in the INS response are possible without a full analysis. The in-plane part, $\Delta_{ab}(k) = \Delta_{1}(\cos(k_{x}) + \cos(k_{y})) + \Delta_{2} \cos(k_{y}) \cos(k_{x})$, of our proposed gap function is shown in Fig. 1. With electron- and hole Fermi sheets well separated as in LDA (or LDA+DMFT), no in-plane gap nodes are possible, in agreement with a host of measurements [1,8,17]. Interestingly, this leads to $\Delta(k + Q) < 0$ for $k$ along $(0,0) - (\pi,\pi)$ and to $\Delta(k + Q) > 0$ for $k$ near $(\pm \pi/2, 0), (0, \pm \pi/2)$. This implies, following earlier work [12], that INS measurements will show appearance of a sharp collective “spin exciton” mode in the U-SC state at $k = (\pi,\pi)$, but none for $k = (\pm \pi/2, 0)$. Of course, incoherent features coming from DMFT propagators will introduce damping of this mode, but the qualitative feature should survive. Since an out-of-plane $\cos(k_{z})$ component is induced in the full gap function due to interband proximity [18] effect, the INS intensity should also reflect this modulation in $q_{z}$. This last prediction is a consequence of our form of the full gap function, and goes beyond previous work [12]. Such a resonance, albeit sizably damped, is indeed seen in INS work on the 122-FePn [10]. Moreover, the $\cos(q_{z})$ form has also been measured by INS on the 122 FePn [11], but remains to be
checked in the 1111 family. Finally, the in-plane angular modulation of the gap function is inferred from \( \mu \text{SR} \) work on the \( \text{Sm-based FePn} \) [4]. Thus, rationalization with extant INS results readily follows directly from our proposal for the gap function.

Next, we discuss the NMR relaxation rate in the normal and U-SC states, making detailed comparison with experimental work. In the normal, incoherent metal state, the reduction in \( T_1^{-1} \) below 200 K [1, 23] indicates opening of a spin gap, as in underdoped cuprates. While the spin gap in cuprates has been identified with short-range magnetic correlations in a quasi-2D, doped quantum antiferromagnet, its origin in the multi-band FePn is not settled. We emphasize that this behavior is quantum antiferromagnet, its origin in the multi-band FePn is not settled. We regard this as a justification for using LDA+DMFT.

NMR work [4] reveals good agreement between theory and experiment around \( x = 0.1 \). The absence of the \( (T/T_1)^{-1} = \text{const} \) regime is striking. In particular, both experiment and our result show a quasi-linear-in-\( T \) (like \( T^{0.8-0.9} \)) increase in \( 1/T_1 \) at “high” \( T > 200 \) K (see inset of Fig. 3). This resembles the high-\( T \) precursor of a quantum critical system, and corresponds to the “strange metal” regime in the \( T \) vs \( x \) phase diagrams for this system [25]. However, as \( T \) is lowered, a smooth drop in \( (T/T_1)^{-1} \) around 150 K marks the onset of the gradual opening up of a spin gap. Given strong frustration \( (J_2/J_1 \sim 0.7) \) in FePn, it is tempting to link this spin gap with strong, short ranged AF correlations, which are expected to survive the doping induced destruction of the SDW [26]. We note that the \( J_1 - J_2 \) model has also been used to provide a quantitative fit of INS results for the undoped 122 FePn, though it is formally valid in the strictly localized regime. This is additional evidence for a strong coupling picture, since, in the itinerant picture, melting of the SDW should yield a paramagnetic Fermi liquid at low \( T \) with no spin gap, at variance with observations. From our results, we estimate a renormalized spin gap scale \( O(150) \) K in the 1111 FePn.

In Fig. 2 we show the NMR \( (T_1 T)^{-1} \) as a function of electron doping for \( \text{LaO}_{1-x} \text{FeAsF}_x \), with \( x = 0.0, 0.1, 0.2 \). Since we do not consider the \( q = (\pi, 0) \) SDW phase, the \( x = 0.0 \) curve should only be trusted above \( T_N = 135 \) K (shown by the black curve in Fig. 2). With \( x = 0.1, 0.2 \), however, SDW order is destroyed, and U-SC emerges at low \( T \). In this range of \( x \), our results can validly be compared to experiment, which we now turn to do.

Quite remarkably, a direct comparison with published}

In fact, \( T_1^{-1} \approx \tanh(0.42T) \) over almost the whole range from low-(\( T > 15 \) K) to high \( T \). While this is not particularly illuminating, it shows that the “marginal” form, \( \chi_{\text{loc}}^\mu(\omega) \sim -\left(\omega/T\right) \), is recovered only at high \( T \), and is cut off by the spin gap around 200–250 K. This bears a peculiar resemblance to underdoped cuprates. However, at very low \( T \), a power-law form \( T_1^{-1} \approx T^{1.5-1.6} \), is seen. This is intriguing, and is fit neither by self-consistent renormalization theory [28], nor by any known local non-

![Fig. 2](image_url)  \( I/T_1 \) (arb. units) vs. \( T (K) \) for various doping levels in \( \text{LaO}_{1-x} \text{FeAsF}_x \). The line styles correspond to different doping levels: \( x = 0.0 \) (black, solid), \( x = 0.1 \) (red, dotted), \( x = 0.2 \) (blue, dot-dashed), and \( x = 0.1 \) (green, dashed). The inset shows a log-log plot of the \( I/T_1 \) vs. \( T \) dependence for different doping levels.

![Fig. 3](image_url)  Low \( T \) behavior of the NMR \( T_1^{-1} \) on a log-log plot (main panel) and on a normal scale (inset). Clear power-law behavior without the Hebel-Slichter coherence peak, in good agreement with experiment [4], is seen.

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**Notes:**
- \( \mu \text{SR} \) refers to the use of muon spin rotation spectroscopy.
- \( T_N = 135 \) K is a critical temperature, above which SDW order is destroyed.
- SDW refers to spin density wave order.
- U-SC stands for undoped superconducting state.
- \( J_1 - J_2 \) model is a simplified model for the interactions in Fe-based superconductors.
- \( \tanh \) is the hyperbolic tangent function.
- \( \chi_{\text{loc}}^\mu(\omega) \) is the local magnetic susceptibility.

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**References:**
- [1] 
- [23] 
- [25] 
- [26] 
- [28]
FL exponents \[29\]. It could involve several, frustrated, nearly degenerate spin fluctuation channels coming from the multiband nature of FePn, but we are unable to quantify this further.

However, we can still make a few qualitative remarks to get more insight. In the strongly correlated metal, with a very small “coherent” component in the DMFT spectral functions \[2, 18\], “Mottness” underpins the low-energy physics. More precisely, when one is close to a correlation-driven Mott insulator, the metallic state has small density of quasi-itinerant carriers co-existing with effectively local moments \[2\]. These latter arise from integrating out the high energy Hubbard bands in the DMFT spectral function, as argued by Baskaran, Si et al. and Wu et al. Given the frustrated hoppings characteristic of FePn, the spin degrees of freedom are qualitatively described by an effective frustrated \(J_1 - J_2\) Heisenberg-type model. In this model, there is a large window in \(T\), between \(T_{SDW}\) and \(T_c\) \[20\], where lattice translational symmetry is spontaneously broken but the spin rotational \((SU(2))\) symmetry is not. This naturally leads to generation of a spin gap, in agreement with observations. Of course, as LDA+DMFT shows, the actual situation in FePn is somewhat removed from a strictly localized limit where the \(J_1 - J_2\) model would apply. However, in view of the Mottness, we believe that it still provides a qualitative understanding of the features derived above in the full DMFT calculation.

At \(T_c\), there is no HS peak, as seen in Fig. 20 in our work, this arises from strong inelastic scattering in the “normal” incoherent state \[18, 20\] (notice that \(\Sigma^\omega_b\) enters the DMFT equation for \(G_b(\omega)\) in the SC state, producing strong damping). At very low \(T < < T_c\), \(T^{-1}_1(T)\) shows a power-law-in-\(T\) dependence: \(T^{-1}_1(T) \sim T^n\), with \(n = 2.2 - 2.5\), qualitatively consistent with observations in the 1111 FePn \[3\], which show neither a \(T^3\) nor a \(T^5\) law for \(T < < T_c\). The two-step variation of \(T^{-1}_1\) below \(T_c\) is also reproduced theoretically. The first “step” from \(T_c > T > T_c/4\) is dominantly governed by the larger gap component, while the lower-\(T\) variation comes from the smaller gap component, as expected from an in-plane anisotropic gap, while the power-law variation is ascribed to out-of-plane line nodes in such a gap. It is still possible that disorder effects (which must be treated in the unitary limit \[1\]) will lift the out-of-plane gap nodes, as discussed by Maier et al. \[12\] and give \(T^{-1}_1 \sim T^3\) behavior \[4\]; this remains to be checked. In our theory, the power law behavior arises from the out-of-plane line nodes, induced in the gap by an interband proximity effect. Since it does not require disorder effects, our conclusion should be more “universal” \[10\]. Thus, our results show how good agreement with the NMR data is derived in the whole \(T\) range in terms of our theoretical picture of an U-SC with proximity induced line nodes, arising from an incoherent normal state at \(T_c\).

In conclusion, we have studied the magnetic fluctuations in FePn, based on a novel theoretical proposal for the symmetry of the SC gap function. In a picture where U-SC with out-of-plane gap nodes arises from an incoherent, strongly correlated normal state, we have shown how the \(T\)-dependence of the NMR relaxation rate can be nicely understood over the whole \(T\) range, from the lowest- to “high” \(T\). Moreover, we have argued how the specific form of the gap function allows for concrete predictions concerning the observation of the collective resonant peak in INS measurements. Our study provides further support for the strongly correlated nature of FePn above \(T_c\), and puts our theoretical proposal of an U-SC with out-of-plane gap nodes on a firmer footing.

[1] For a review of the controversy over the pair symmetry, see I.I. Mazin and J. Schmalian, arXiv:0901.4790
[2] Q. Si and E. Abrahams, Phys. Rev. Lett. 101, 076401 (2008); ibid, J. Wu et al., Phys. Rev. Lett. 101, 126401 (2008); G. Baskaran, J. Phys. Soc. Jpn. 77, 113713 (2008); Q. Si et al., arXiv:0901.4112
[3] H. He et al., Phys. Rev. Lett. 86, 1610 (2001); ibid. J. Bobrow et al., Phys. Rev. Lett. 78, 3757 (1997).
[4] Y. Nakai et al., arXiv:0810.3569.
[5] M.E. Zhitomirsky and T.M. Rice, Phys. Rev. Lett. 87, 057001 (2001).
[6] D. Parker et al., Phys. Rev. B 78, 134524 (2008).
[7] S. Graser et al., New J. Phys. 11, 025016 (2009); and, V. Mishra et al., 0901.2653.
[8] L. Wray et al., Phys. Rev. B 78, 184508 (2008).
[9] L. Malone et al., arXiv:0806.3908.
[10] A. D. Christianson et al., Nature, 456, 930 (2008).
[11] S. Chi et al., arXiv:0812.1351.
[12] M.M. Korshunov and I. Eremin, Phys. Rev. B 78, 140509(R) (2008); T.A. Maier et al., arXiv: 0805.0316; ibid arXiv: 0903.0008.
[13] G. M. Zhang et al., arXiv:0809.3874.
[14] J. Wu and P. Phillips, arXiv:0901.3538.
[15] J.D. Fletcher et al., arXiv:0812.3858.
[16] R. Prozorov et al., arXiv:0901.3698.
[17] T. Y. Chen et al., Nature 453, 1224 (2008).
[18] M.S. Laad and L. Craco, arXiv:0902.3400.
[19] K. Haule et al., Phys. Rev. Lett. 100, 226402 (2008).
[20] L. Craco et al., Phys. Rev. B 78, 134511 (2008); ibid M. S. Laad et al., Phys. Rev. B, 79, 024515 (2009).
[21] J. Tahir-Kheli, Phys. Rev. B 58, 12307 (1998).
[22] U. Chatterjee et al., Phys. Rev. B 75, 172504 (2007).
[23] We compare our theory with the results of Ref. \[4\]. For more work, see S. Kawasaki et al., Phys. Rev. B, 78, 221506(R) (2008); Y. Nakai et al., J. Phys. Soc. Jpn. 77, 073701 (2008).
[24] A.V. Boris et al., Phys. Rev. Lett. 102, 027001 (2009); ibid S.I. Mirzaei et al., arXiv:0806.2303.
[25] C. Hess et al., arXiv:0811.1601.
[26] C. Xu et al., Phys. Rev. B 78, 020501(R) (2008).
[27] S.O. Diallo et al., arXiv:0901.3784.
[28] T. Moriya and K. Ueda, Rep. Prog. Phys. 66, 1299 (2003).
[29] K. Ingersent and Q. Si, Phys. Rev. Lett. 89, 076403 (2002).