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Theory of magnetic fluctuations in iron pnictides

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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, tracking their evolution from the incoherent normal, pseudogapped metal, to the U-SC state. Within our proposal of extended-s-plus s_{xy} (generalized $s_{\pm}$) symmetry of the inplane gap component 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 in good agreement with experiment, and (ii) we predict a sharp resonance in the U-SC state along (π,π), 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.

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I. INTRODUCTION

The precise mechanism of unconventional superconductivity (U-SC) in the recently discovered iron pnictides (FePn) is presently a hotly debated issue.[1] While many physical responses are reminiscent of cuprates,[2] 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.

Study of magnetic fluctuations in an U-SC can help unearth the symmetry of the SC order parameter, as shown by detailed studies for cuprates.[3] In the FePn, NMR studies already reveal normal state pseudogap behavior[4] and U-SC. The spin-lattice relaxation rate, T_1^{-1}, shows marked deviation from the linear-in-T Korringa form expected from a Fermi liquid and smoothly *decreases* for $T<2T_c$, where T_c is the SC transition temperature. At very low T, one finds $T_1^{-1}\simeq T^n$ with $n\simeq 2.3-2.5$, indicating line nodes in the SC gap. However, other probes reveal anisotropic, albeit fully gapped, structure of the in-plane gap function. Thus, extant data imply that, either one has out-of-plane line nodes, as in Sr_2RuO_4,[5] or disorder effects in a $s_{\pm}$-SC produce the observed behavior.[6] In the extended-s wave idea, disorder is argued to lift the nodal structure, again giving similar behavior.[7] The issue is thus controversial: while angle-resolved photoemission spectra (ARPES) data are inconclusive regarding existence of nodes on electronlike Fermi sheets (FSs),[8] a penetration depth study, at least in $SmO_{1-x}FeAsF_x$, shows *smooth* angular variation of the in-plane gap.[9] While inelastic neutron scattering (INS) work does reveal a low-energy resonance structure in the U-SC state for $\mathbf{Q}=(\pi,\pi)$,[10] more detailed map of the INS response in $\mathbf{q}$ space awaits future work. To date, we are aware of one study where the dynamical spin susceptibility, $\chi''(\mathbf{q},\omega)$, 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.[11]

Extant theoretical works have studied these issues using effective model Hamiltonians, both in the weak[12] and strong[13,14] coupling limits. In the itinerant approach, the magnetic fluctuations have been computed within HF-RPA. For $s_{\pm}$ pairing, a sharp resonance for $\mathbf{Q}=(\pi,\pi)$ in INS is predicted below T_c,[12] while no such feature is arises for s, ex-s, and d wave-pairing, or for $\mathbf{q}\neq\mathbf{Q}$. To get the power-law-in-T behavior in NMR and μSR 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, $\lambda(T)\simeq T^{1.2}$,[15] 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.[16] In the strong coupling limit, an *effective* model description based on a reduced two-band model has been proposed.[17] This effective $t-J_1-J_2$ model has also been solved within the HF-RPA approximation, and, once more, a spin resonance as a unique fingerprint of the $s_{\pm}$ pair symmetry is deduced. Additionally, the RPA correction to $1/T_1$ is computed to be qualitatively in good agreement with experiment, but only in the U-SC state. But the anomalous normal state magnetic responses as seen in NMR remain to be investigated in detail within intermediate-to-strong coupling approaches. The anomalies in $1/T_1$ mentioned above are, however, out of scope of the weak-coupling HF-RPA. This is because, in the doped regime without SDW order, destruction of the SDW reverts the system to an itinerant FL, which, by construction, can only exhibit a Korringa law above T_c, in stark conflict with observations. Thus, a unified understanding of the evolution of the NMR relaxation rate over the whole temperature range, from the high-T (incoherent metal) to the low-T (U-SC) regime in doped FePn in a *single* theoretical picture remains an interesting, unresolved issue.

In particular, the observation of a normal state pseudogap in $1/T_1$, power-law behavior for $T\ll T_c$, and absence of the Hebel-Slichter peak just below T_c severely constrain theoretical models for the FePn. Taken together, as they must be, these imply that an incoherent, spin pseudogapped and incoherent "normal" state evolves into an U-SC in doped FePn as T is lowered. Further constraints are imposed by ARPES (Ref. 8) and tunnelling data,[18] which show no in-plane nodes in the SC gap function in FePn. *If* this is true, one must consider both the nuclear magnetic resonance (NMR) and INS data within a theoretical scenario with possible out-of-plane line nodes in the SC gap.

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 (the generalized $s_{\pm}$ symmetry, $\Delta_{plane}(k)=\Delta_1(c_x+c_y)+\Delta_2 c_x c_y$ with $c_\alpha=\cos(k_\alpha)$, where $\alpha=x,y$ and $\Delta_2/\Delta_1=0.7-1.0$, so with no in-plane nodes) *and* inter-band proximity induced out-of-plane line nodes.[19] In contrast to the itinerant picture, our proposal, akin to that proposed by Haule *et al.*[20] and us[21,22] for the "normal" phase, is based on a strong correlation view of FePn. In earlier work, we have shown how our intermediate-to-strong coupling picture leads to *quantitatively* good agreement with a host of basic physical properties of the doped 1111-FePn in *both* the normal as well as the U-SC phases. In our modeling, normal state incoherence arises from the proximity of the FePn to a Mott insulator.[2,19–22] Here, we investigate the NMR response within such a correlated approach in detail, using the *full*, multiband spectral functions for *all d* orbitals. LDA+DMFT can readily access the intermediate coupling regime relevant for FePn.[20] 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, in qualitative accord with INS results.

II. THEORY AND RESULTS

The central quantity of interest is the dynamical spin susceptibility, $\chi(\mathbf{q},\omega)=\Sigma_{a,b}\chi_{ab}(\mathbf{q},\omega)$, where a,b are *all* d-orbital indices, and $\mathbf{q},\omega$ are the momentum and energy transfers in INS. Viewing FePn as strongly correlated systems with $U=4.0$ eV, $U'=2.6$ eV and $J_H=0.7$ eV, we construct $\chi(\mathbf{q},\omega)$ in terms of the *full* LDA+DMFT propagators computed in earlier work.[19,21,22] Very 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. It is thus very interesting to inquire if the *same* approach can also quantitatively describe the evolution of magnetic fluctuations in the doped FePn with T. In this paper, we describe how this can indeed be done.

Our 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, multiorbital (MO) correlations is included from the outset. For a MO-system, after replacing the *bare* $G_{aa}(\mathbf{k},\omega)$ with $G_{aa}(\mathbf{k},\omega) \equiv G_{aa}^{LDA+DMFT}(\mathbf{k},\omega)=[\omega-\epsilon_{ka}-\Sigma_a(\omega)-\frac{\Delta_{ab}^2(k)}{\omega+\epsilon_{kb}+\Sigma_b^*(\omega)}]^{-1}$ and $F_{ab}(k,\omega)=G_{aa}(k,\omega)\frac{\Delta_{ab}(k)}{\omega+\epsilon_{kb}+\Sigma_b^*(\omega)}$, and introducing the spin operator $S_{a,\mu}(\mathbf{q})=\frac{1}{2}\Sigma_{\mathbf{k}} c_{a,\mu,\sigma}^\dagger(\mathbf{k}+\mathbf{q})\sigma_{a,\sigma,\sigma'}^\mu c_{a,\mu,\sigma'}(\mathbf{k})$, with $\mu=x,y,z$, the "bare" dynamical spin susceptibility reads

$$\chi_{0,a,b}^{\mu\nu}(\mathbf{q},\omega) = -\frac{1}{2}\sigma_{a,\sigma\sigma'}^\mu \cdot \sigma_{b,\sigma\sigma'}^\nu \sum_{\mathbf{k},\omega'} \times[G_{aa}(\mathbf{k}+\mathbf{q},\omega+\omega')G_{bb}(\mathbf{k},\omega') + F_{ab}(-\mathbf{k}-\mathbf{q},-\omega-\omega')F_{ba}(\mathbf{k},\omega')].$$

We emphasize that $\chi_{0,a,b}^{\mu\nu}(\mathbf{q},\omega)$ has *both*, intra- and inter-orbital components. Including the ladder vertex in an infinite summation of "ladder" diagrams using RPA, the renormalized magnetic susceptibility, $\chi_{a,b}(\mathbf{q},\omega) =[\chi_{0,a,b}^{-1}(\omega)-J(\mathbf{q})]^{-1}$, where $\chi_{0,a,b}(\omega)=\Sigma_{\mathbf{q}}\chi_{0,a,b}(\mathbf{q},\omega)$ and $J(\mathbf{q})=J_1(\cos(q_x a)+\cos(q_y a))+J_2\cos(q_x a)\cos(q_y a)$, with $J_1 \simeq \frac{t_{ab}^2}{U'+J_H}$ and $J_2 \simeq \frac{t_{ab}'^2}{U'+J_H}$ being the frustrated superexchange scales in FePn.[2] Using $\chi_{0,a,b}(\epsilon)=C\int d\epsilon f(\epsilon)[1-f(\epsilon)]W(\epsilon)$ in the RPA series, the NMR relaxation rate, $T_1^{-1}=\Sigma_{\mathbf{q}}\frac{\chi''(\mathbf{q},\omega)}{\omega}|_{\omega\to 0}$, can be now expressed in terms of the *full* DMFT propagators. Here, $W(\epsilon) =\Sigma_{a,b}[\rho_{aa}(\epsilon)\rho_{bb}(\epsilon)+\rho_{ab}(\epsilon)\rho_{ba}(\epsilon)]$ [23] and the $\rho_{aa}(\epsilon),\rho_{ab}(\epsilon)$ are the LDA+DMFT local spectral functions computed earlier.[19] Also, $C=2(\frac{2\pi}{\hbar})(\gamma_e\gamma_n\hbar)^2\langle\frac{1}{r^3}\rangle$. Finally, our restriction to the noncrossing diagrams in the ladder summation for $\chi(\mathbf{q},\omega)$ is an approximation. It is possible that "noncrossing" diagrams need to be included in a full description. However, for the underdoped cuprates, it has been shown that a renormalized "RPA" summation for $\chi(\mathbf{q},\omega)$ with fully renormalized one-particle $G_\sigma(k,\omega),F_{\sigma,-\sigma}(k,\omega)$ gives excellent reconciliation of ARPES and INS data.[24] 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 $T_1^{-1}\simeq e^{-\Delta/kT}$ survives for $T\ll T_c$ 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 $F_{ab}(\mathbf{k},\omega)=0$. 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, $\Delta_{ab}(k)=\Delta_1(\cos(k_x a)+\cos(k_y a)) +\Delta_2\cos(k_x a)\cos(k_y a)$, of our proposed gap function[19] is shown in Fig. 1. With electron- and hole Fermi sheets well

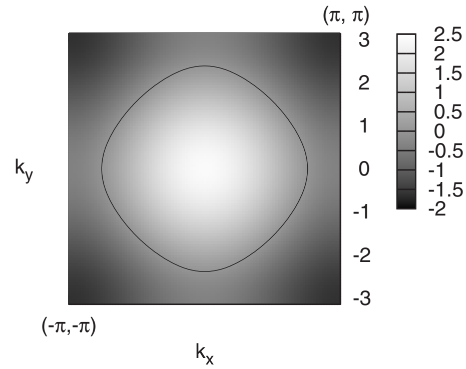

FIG. 1. Proposed gap function for the 1111-Iron Pnictides. The gap function has nearest-(Δ_1, with ex-s symmetry) and next-nearest (Δ_2, with s_{xy} symmetry) neighbor components. With $\Delta_2/\Delta_1=0.375$, no in-plane gap nodes arise in the gap function, in agreement with experiment (Ref. 8).

separated as in LDA (or LDA+DMFT), no in-plane gap nodes are possible, in agreement with a host of measurements.[1,8,18] Interestingly, this leads to $\Delta(\mathbf{k}+\mathbf{Q})\Delta(\mathbf{k})<0$ for $\mathbf{k}$ along $(0,0)-(\pi,\pi)$ and to $\Delta(\mathbf{k}+\mathbf{Q})\Delta(\mathbf{k})>0$ for $\mathbf{k}$ 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 $\mathbf{k}=(\pi,\pi)$, but none for $\mathbf{k}=(\pm\pi/2,0)$. 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 $\cos(k_z c)$ component is induced in the full gap function due to interband proximity effect,[19] 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 $\cos(q_z c)$ form is consistent with INS measurements on 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 μSR work on the Sm-based FePn.[9] 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[4,25] 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 multiband FePn is not settled. We emphasize that this behavior is observed in the same regime where PES[8] data show *incoherent* charge dynamics, corroborated by a linear-in-T resistivity, a T-dependent Hall constant, and no Drude peak in optics.[26] All these are compelling indicators of a strongly correlated metal. We regard this as a justification for using LDA+DMFT.

In Fig. 2, we show the NMR $(T_1T)^{-1}$ as a function of electron doping for $LaO_{1-x}FeAsF_x$, with $x=0.0,0.1,0.2$. Since we do not consider the $\mathbf{q}=(\pi,0)$ SDW phase, the $x=0.0$ curve should only be trusted above $T=T_N=135$ 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 NMR work[4] reveals good agreement between theory and experiment around $x=0.1$. The absence of the $(T_1T)^{-1}=const$ 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.[27] However, as T is lowered, a *smooth* drop in $(T_1T)^{-1}$ around 150 K marks the onset of the gradual opening up of a *spin* gap. Given strong frustration $(J_2/J_1\simeq 0.7)$ 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.[28] We note that the J_1-J_2 [29] model has also been used to provide a

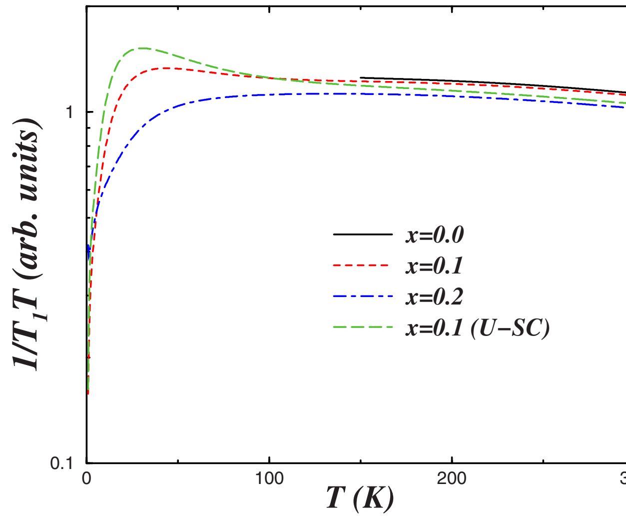

FIG. 2. (Color online) T-dependence of the NMR $(T_1T)^{-1}$ over the full T range for $LaO_{1-x}FeAsF_x$, with $x=0$ (black), $x=0.1$ (red, dotted), $x=0.2$ (blue, dot-dashed) and with inclusion of U-SC for $x=0.1$ (green, dashed). Notice how a doping-dependent spin gap around $T^*\simeq 150$ K opens in the doped case ($x=0.1,0.2$), in good agreement with experiment (Ref. 4).

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 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, $\chi''_{loc}(\omega)\simeq-(\omega/T)$, 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.[3] However, at very low

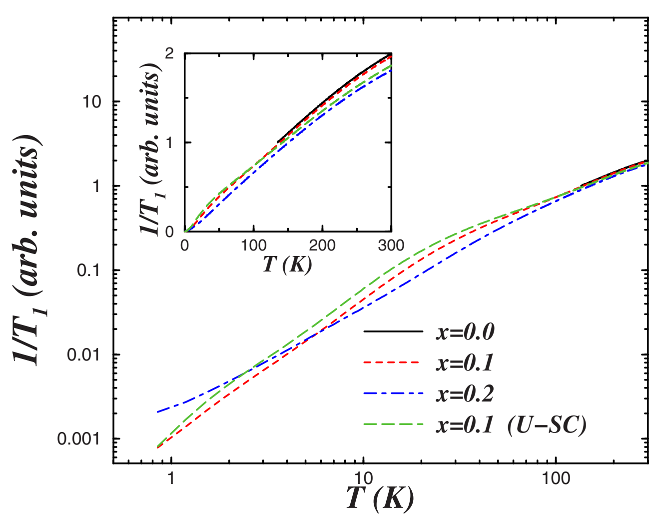

FIG. 3. (Color online) Low T behavior of the NMR T_1^{-1} on a log-log plot (main panel) and on a normal scale (inset). Clear power-law behavior without the Hebel-Slichter coherence peak, in good agreement with experiment (Ref. 4), is seen.

T, a power-law form $T_1^{-1} \simeq T^{1.5-1.6}$, is seen. This is intriguing, and is fit neither by self-consistent renormalization theory,[30] nor by any known local non-FL exponents.[31] 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,[2,19] "Mottness" underpins the low-energy physics. 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_s,[28] 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 *low* energy features derived above in the full DMFT calculation.

At T_c, there is no HS peak, as seen in Fig. 3: in our work, this arises from strong inelastic scattering in the "normal" incoherent state[19,21,22] (notice that $\Sigma_b^*(\omega)$ enters the DMFT equation for $G_a(\omega)$ in the SC state, producing strong damping). At very low $T \ll T_c$, $T_1^{-1}(T)$ shows a power-law-in-T dependence: $T_1^{-1}(T) \simeq T^n$, with $n=2.2-2.5$, qualitatively consistent with observations in the 1111 FePn,[4] which show neither a T^3 nor a T^5 law for $T \ll T_c$. 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 $T_1^{-1} \simeq T^3$ behavior;[6] 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."[16] 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.

III. DISCUSSION

Our approach is very different from those hitherto considered. Weak coupling approaches[6,7] *cannot*, by construction, access the non-Korringa and pseudogap features in $1/T_1$ in the normal state. In the $t-J_1-J_2$ modeling, the normal state anomalies in $1/T_1$ have not been considered. In any case, dHvA results on LaFePO suggest that the FePn are somewhat removed from a strictly localized regime where such a $t-J_1-J_2$ modeling might be expected to hold. In our approach, proximity to a correlation-driven Mott insulator implies that the physical responses in FePn are controlled by *dualistic* electronic states, as discussed above. A small number of "itinerant" carriers give the carrier pockets observed in ARPES and dHvA data, while the "localized" carriers (residing in the correlation-induced Hubbard bands) at high energy provide local magnetic moments. Given strong geometric frustration in FePn, suppression of the $\mathbf{q}=(\pi,0)$ magnetic order by doping still leaves short-ranged, frustrated spin correlations intact. It is precisely these correlations which result in a spin pseudogap observed in the NMR relaxation rate in the "normal" state of the 1111-FePn. Additionally, in the SC state, lack of the Hebel-Slichter peak and power-law T-dependence of $1/T_1$ strongly support possible gap nodes in the U-SC state. We have shown how *all* these "strange" features can be (even semiquantitatively) be understood within our specific proposal. The central message of our approach is thus that an intermediate-to-strong coupling picture of the 1111-FePn leads to a satisfying description of the evolution of local spin fluctuations with T over the whole range of interest, from high-(incoherent metal) to low-T (U-SC). Clearly, being based on a first-principles correlated (LDA+DMFT) approach involving the *full* multiband electronic structure of the FePn, our proposal goes way beyond earlier theoretical work (see below). In fact, in the intermediate coupling regime, an itinerant-localized duality underpins the physical behavior of the system, simultaneously giving rise to *bands* as observed in dHvA studies, and to dynamically fluctuating local momemts, as measured in NMR, putting FePn close to "Mottness."

Finally, let us make a few comments putting our work in perspective in the light of earlier studies. First, we notice that LDA calculations claim[32] to find good "agreement" with ARPES data. Upon a closer scrutiny, one finds that the LDA bands need to be shifted by orbital-dependent amounts *and* narrowed by a factor of 2–3 to achieve a reasonable fit with ARPES. On the other hand, INS dispersions have been accounted for by low-order spin-wave calculation for a Heisenberg model[33] with appreciable frustration. Even though the spin-wave *dispersion* is reproduced well thereby, we opine, since dynamical fluctuations are ignored, that the ω-dependent INS lineshape will not be well reproduced. In fact, examination of the normal state INS intensity clearly shows appreciably broad lineshapes. It is very difficult, if not impossible, to discern long-lived, propagating spin-wave modes, though a (broadened) resonance indeed develops below T_c, broadly consistent with requirements of ex-s pair symmetry.

These observations clearly imply the necessity to go beyond a weakly correlated band description, since an effective local moment picture underlying the use of the Heisenberg-like model itself *cannot* be consistently obtained within a band description. Clearly, both the above effects are direct fall-outs of correlations: the MO Hartree (static, but orbital-dependent) corrections shift the LDA bands by varying amounts, and the band-narrowing is a direct consequence of dynamical correlations. LDA+DMFT indeed captures *both* these effects adequately for the 1111-FePn, as shown in earlier work.[20–22] Obviously, if description of the one-particle spectrum requires such correlation effects, it is only natural that these should be important for the description of experiments probing collective charge- and spin fluctuations. Generation of *effective* local moments is a natural consequence of a metallic system close to Mottness, where carriers have a dualistic (itinerant-localized) character, as is ubiquitous to correlated matter. A subsequent description of spin fluctuations within an *effective* Heisenberg picture is then possible. This cannot be understood in a band picture, where SDW arises from FS nesting: in such a picture, the spin-waves will look very different from those derived from a Heisenberg-like model, and no spin pseudogap behavior is expected in NMR. These constitute our motivations for employing LDA+DMFT to treat sizable d-band correlations in the FePn. Of course, truly *ab-initio* values of U,U' used in our LDA+DMFT, or in any comparable method, are not known. We have employed values close to those estimated by Haule *et al.*,[20] believed to be reasonable for Fe-d shells in FePn.

Based on good quantitative agreement between our LDA+DMFT results and NMR data, we propose that magnetic fluctuations are much better accessed by use of the LDA+DMFT propagators, rather than the free electron (band) Green's functions, since they include sizable dynamical correlations that characterize the intermediate correlation case (severely underestimated by LDA+HF-RPA and overestimated by purely localized spin models). Interestingly, our intermediate coupling scheme based on "Mottness" is able to capture the T-dependence of $1/T_1T$ in the whole temperature range, from the "normal" incoherent state, to the power-law form well below T_c. These features arise from the incoherent spectral functions (due to sizable d-band correlations) obtained within LDA+DMFT as shown here. This is the main message of our work. If, on the other hand, one uses L(S)DA or L(S)DA+U spectral functions, the following is expected, based upon very general and valid argumentation: since LDA or LDA+U spectral functions, by construction, represent coherent (in the sense of FL) band states, the NMR response expected therefrom must correspond to that of a FL, i.e, $1/T_1$ must follow a Korringa-like, linear-in-T dependence, with no spin pseudogap signatures. No other T-dependence is possible, since the FePn are metallic, and LDA+U will not produce local moments (which could, in principle, show unusual behavior, but this does not hold for the FeFn with LDA+U). Indeed, we have checked that using small $U(\approx 1.0$ eV), the Green's functions are FL like (i.e, they have a sizable quasiparticle pole in the density of states), and that using these does not describe the PES/XAS or optical responses[22] in the 1111-pnictides. Our results show that a sizable $U=4.0$ eV, along with $J_H=0.7$ eV,[21,22] is required to achieve a consistent description of both PES/XAS and optics in the normal state. In previous works,[34] the incoherent excitations in the normal state giving non-Korringa and pseudogap behavior in NMR relaxation in the normal state are missed. Using a Green's function with poles, as in these works (or in the band picture) cannot produce these behaviors. In our work, the power-law behavior below T_c does not come from disorder, but due to possible multiband proximity induced c-axis nodes in the gap function. Hence, these features should be rather universal, as indeed inferred from μSR studies.[16] Our picture is thus very different from extant proposals, and does not conflict other observations, such as tunnelling,[18] which show no in-plane nodes in the superconducting gap function.

Such incoherent features are seemingly common to other Fe-based SCs as well: in $FeSe_{1-x}$,[35] $1/T_1$ shows an even stronger non-FL T-dependence in the "normal" state, and U-SC develops directly from an insulator-like incoherent state at lower T. In this light, extensions of our approach here to the FeSe family should be readily applicable: we leave this for a future work.

IV. CONCLUSION

In conclusion, we have studied the magnetic fluctuations in FePn, based on a 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.

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[1] For a review of the controversy over the pair symmetry, see I. I. Mazin and J. Schmalian, Physica C **469**, 614 (2009).

[2] Q. Si and E. Abrahams, Phys. Rev. Lett. **101**, 076401 (2008); J. Wu, P. Phillips, and A. H. Castro Neto, *ibid.* **101**, 126401 (2008); G. Baskaran, J. Phys. Soc. Jpn. **77**, 113713 (2008); Q. Si, E. Abrahams, J. Dai, and J.-X. Zhu, New J. Phys. **11**, 045001 (2009).

[3] H. He, Y. Sidis, P. Bourges, G. D. Gu, A. Ivanov, N. Koshizuka, B. Liang, C. T. Lin, L. P. Regnault, E. Schoenherr, and B. Keimer, Phys. Rev. Lett. **86**, 1610 (2001); J. Bobroff, H. Alloul, P. Mendels, V. Viallet, J.-F. Marucco, and D. Colson, *ibid.* **78**, 3757 (1997).

[4] Y. Nakai, S. Kitagawa, K. Ishida, Y. Kamihara, M. Hirano, and H. Hosono, Phys. Rev. B **79**, 212506 (2009).

[5] M. E. Zhitomirsky and T. M. Rice, Phys. Rev. Lett. **87**, 057001 (2001).

[6] D. Parker, O. V. Dolgov, M. M. Korshunov, A. A. Golubov, and I. I. Mazin, Phys. Rev. B **78**, 134524 (2008).

[7] S. Graser, T. A. Maier, P. J. Hirschfeld, and D. J. Scalapino, New J. Phys. **11**, 025016 (2009); V. Mishra, G. Boyd, S. Graser, T. Maier, P. J. Hirschfeld, and D. J. Scalapino, Phys. Rev. B **79**, 094512 (2009).

[8] L. Wray, D. Qian, D. Hsieh, Y. Xia, L. Li, J. G. Checkelsky, A. Pasupathy, K. K. Gomes, C. V. Parker, A. V. Fedorov, G. F. Chen, J. L. Luo, A. Yazdani, N. P. Ong, N. L. Wang, and M. Z. Hasan, Phys. Rev. B **78**, 184508 (2008).

[9] L. Malone, J. D. Fletcher, A. Serafin, A. Carrington, N. D. Zhigadlo, Z. Bukowski, S. Katrych, and J. Karpinski, Phys. Rev. B **79**, 140501(R) (2009).

[10] A. D. Christianson, E. A. Goremychkin, R. Osborn, S. Rosenkranz, M. D. Lumsden, C. D. Malliakas, I. S. Todorov, H. Claus, D. Y. Chung, M. G. Kanatzidis, R. I. Bewley, and T. Guidi, Nature (London) **456**, 930 (2008).

[11] S. Chi, A. Schneidewind, J. Zhao, L. W. Harriger, L. Li, Y. Luo, G. Cao, and Zhu'an Xu, M. Loewenhaupt, J. Hu, and P. Dai, Phys. Rev. Lett. **102**, 107006 (2009).

[12] M. M. Korshunov and I. Eremin, Phys. Rev. B **78**, 140509(R) (2008); T. A. Maier and D. J. Scalapino, *ibid.* **78**, 020514(R) (2008); T. A. Maier, S. Graser, D. J. Scalapino, and P. Hirschfeld, *ibid.* **79**, 134520 (2009).

[13] G.-M. Zhang, Y.-H. Su, Z.-Y. Lu, Z.-Y. Weng, D.-H. Lee, and T. Xiang, EPL **86**, 37006 (2009).

[14] J. Wu and P. Phillips, arXiv:0901.3538 (unpublished).

[15] J. Fletcher, A. Serafin, L. Malone, J. Analytis, J. Chu, A. Erickson, I. Fisher, and A. Carrington, Phys. Rev. Lett. **102**, 147001 (2009).

[16] R. Prozorov, M. A. Tanatar, R. T. Gordon, C. Martin, H. Kim, V. G. Kogan, N. Ni, M. E. Tillman, S. L. Bud'ko, and P. C. Canfield, Physica C **469**, 582 (2009).

[17] K. Seo, C. Fang, B. A. Bernevig, and J. Hu, Phys. Rev. B **79**, 235207 (2009).

[18] T. Y. Chen, Z. Tesanovic, R. H. Liu, X. H. Chen, and C. L. Chien, Nature (London) **453**, 1224 (2008).

[19] M. S. Laad and L. Craco, Phys. Rev. Lett. **103**, 017002 (2009).

[20] K. Haule, J. H. Shim, and G. Kotliar, Phys. Rev. Lett. **100**, 226402 (2008).

[21] L. Craco, M. S. Laad, S. Leoni, and H. Rosner, Phys. Rev. B **78**, 134511 (2008).

[22] M. S. Laad, L. Craco, S. Leoni, and H. Rosner, Phys. Rev. B **79**, 024515 (2009).

[23] J. Tahir-Kheli, Phys. Rev. B **58**, 12307 (1998).

[24] U. Chatterjee, D. K. Morr, M. R. Norman, M. Randeria, A. Kanigel, M. Shi, E. Rossi, A. Kaminski, H. M. Fretwell, S. Rosenkranz, K. Kadowaki, and J. C. Campuzano, Phys. Rev. B **75**, 172504 (2007).

[25] We compare our theory with the results of Ref. 4. For more work, see K. Ahilan, F. L. Ning, T. Imai, A. S. Sefat, R. Jin, M. A. McGuire, B. C. Sales, and D. Mandrus, Phys. Rev. B **78**, 100501(R) (2008); S. Kawasaki, K. Shimada, G. F. Chen, J. L. Luo, N. L. Wang, and Guo-qing Zheng, Phys Rev. B **78**, 220506(R) (2008); Y. Nakai, K. Ishida, Y. Kamihara, M. Hirano, and H. Hosono, J. Phys. Soc. Jpn. **77**, 073701 (2008).

[26] A. V. Boris, N. N. Kovaleva, S. S. A. Seo, J. S. Kim, P. Popovich, Y. Matiks, R. K. Kremer, and B. Keimer, Phys. Rev. Lett. **102**, 027001 (2009).

[27] C. Hess, A. Kondrat, A. Narduzzo, J. Hamann-Borrero, R. Klingeler, J. Werner, G. Behr, and B. Büchner, Europhys. Lett. **87**, 17005 (2009).

[28] C. Xu, M. Müller, and S. Sachdev, Phys. Rev. B **78**, 020501(R) (2008).

[29] S. O. Diallo, V. P. Antropov, T. G. Perring, C. Broholm, J. J. Pulikkotil, 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] T. Moriya and K. Ueda, Rep. Prog. Phys. **66**, 1299 (2003).

[31] K. Ingersent and Q. Si, Phys. Rev. Lett. **89**, 076403 (2002).

[32] D. H. Lu, M. Yi, S.-K. Mo, A. S. Erickson, J. Analytis, J.-H. Chu, D. J. Singh, Z. Hussain, T. H. Geballe, I. R. Fisher, and Z.-X. Shen, Nature (London) **455**, 81 (2008).

[33] T. Yildirim, Phys. Rev. Lett. **101**, 057010 (2008).

[34] K. Kuroki, H. Usui, S. Onari, R. Arita, and H. Aoki, Phys. Rev. B **79**, 224511 (2009); K. Kuroki, S. Onari, R. Arita, H. Usui, Y. Tanaka, H. Kontani, and Hideo Aoki, Phys. Rev. Lett. **101**, 087004 (2008); K. Kuroki and H. Aoki, Physica C **469**, 635 (2009).

[35] T. Imai, K. Ahilan, F. L. Ning, T. M. McQueen, and R. J. Cava, Phys. Rev. Lett. **102**, 177005 (2009).