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# Universal magnetic and structural behaviors in the iron arsenides

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Commonalities among the order parameters of the ubiquitous antiferromagnetism present in the parent compounds of the iron arsenide high-temperature superconductors are explored. Additionally, comparison is made between the well established two-dimensional Heisenberg-Ising magnet,  $K_2NiF_4$ , and iron arsenide systems residing at a critical point whose structural and magnetic phase transitions coincide. In particular, analysis is presented regarding two distinct classes of phase-transition behavior reflected in the development of antiferromagnetic and structural order in the three main classes of iron arsenide superconductors. Two distinct universality classes are mirrored in their magnetic phase transitions which empirically are determined by the proximity of the coupled structural and magnetic phase transitions in these materials.

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### I. INTRODUCTION

Understanding the fundamental properties of the underlying magnetic and structural orders in the newly discovered iron-based high-temperature superconductors (high  $T_c$ ) is an essential step in the eventual resolution of magnetism's role in the superconducting pairing in these systems. The key unifying feature between the iron arsenide class of high- $T_c$ superconductors and the well-known cuprate high  $T_c$ 's is the universal presence of an antiferromagnetically (AF) ordered state in close proximity to the development of superconductivity within their respective phase diagrams.<sup>1-3</sup> The undoped, parent systems of both classes of high  $T_c$ 's exhibit long-range, AF order<sup>1,4</sup> that is suppressed upon doping and either vanishes<sup>5</sup> or weakly competes<sup>6,7</sup> with the onset of superconductivity. While previous exploration of critical properties intrinsic to the static AF order in the cuprates led to fundamental insights regarding the interactions and physics pertaining to the spin behavior in those systems,<sup>8</sup> experiments probing the detailed behavior of magnetism in the iron pnictide systems are only just beginning.

The ordered spin structures of the LaFeAsO(1111),<sup>4</sup> NaFeAs(111),<sup>9</sup> and BaFe<sub>2</sub>As<sub>2</sub>(122)-type<sup>10</sup> iron arsenide compounds possess a common unidirectional antiferromagnetic ordering as shown in Fig. 1 where the AF propagation vector points both along the long axis within the basal plane and along the out-of-plane, c axis. The in-plane component of the AF wave vector is determined by an accompanying or preceding structural distortion from tetragonal to orthorhombic symmetry in which the *a* axis within the iron layers is slightly elongated. Within the bilayer AFe<sub>2</sub>As<sub>2</sub> class of pnictides (where single crystals are most easily synthesized) a number of early studies suggested first-order behavior in the onset of the AF phase.<sup>11–14</sup> While these reports seemingly precluded any subsequent study of critical behavior in these systems, separate reports also showed continuous, secondorder magnetic phase transitions in a number of the same materials.<sup>15–17</sup> Recently, new single-crystal studies have appeared showing continuous phase transitions in several iron pnictide variants<sup>7,18–20</sup> thereby suggesting that improvements in sample quality and diversity may now facilitate a reliable comparison between materials.

While the exchange couplings and spin dynamics manifest within the iron arsenides have been a topic of considerable focus, relatively little attention has been given to any precise investigation of their magnetic phase-transition behavior. In particular, recent measurements of the magnetic



FIG. 1. (Color online) Crystal structures of representative materials from the (a) REFeAsO, (b) AMFeAs, and (c)  $AFe_2As_2$  classes of iron arsenides. Boxes show the enclosed chemical unit cells of each system. To the right of each chemical structure are plotted the ordered spin systems. Moment directions are plotted as yellow arrows located on the Fe sites.

exchange coupling constants within the parent 122 pnictide class of systems have revealed a drastic modification to the in-plane Fe-Fe exchange energies upon cooling below  $T_S$ .<sup>21</sup> The drastic asymmetry which develops between the  $J_{1A}$  and  $J_{1B}$  exchange parameters upon entering the orthorhombic state necessitates remarkable, simultaneous changes in the phase behavior of the accompanying magnetic order. Examining the magnetic and structural phase behaviors in these 122 compounds, where  $T_S$  and  $T_N$  are coincident, relative to phase behaviors in other pnictide systems, where  $T_S > T_N$ , therefore has the potential to elucidate better the underlying correlations in these systems.

In this paper, we present our analysis of the magnetic and structural order parameters in the three main classes of iron pnictide superconductors:  $AFe_2As_2$  (A site=Ba,Sr,Eu,...), AMFeAs (AM=alkali metal=Na,Li), and REFeAsO (RE =rare earth=La, Pr, Ce,...). Undoped systems within the bilayer, 122 class exhibit a two-dimensional (2D) Ising-like magnetism whose order parameters are shown to mirror closely the spin behavior in the prototypical 2D-Heisenberg-Ising system K<sub>2</sub>NiF<sub>4</sub>. Once the 122 systems are perturbed and the structural and magnetic phase transitions no longer coincide in temperature, a fundamentally different magnetic behavior emerges. In this regime, when  $T_N < T_S$ , a dramatically altered development of the magnetic order parameter appears and collapses onto a seemingly universal curve with a critical exponent of  $\beta \approx 0.25$ . This reflects a crossover from the coincident  $T_N = T_S$ , 2D-Ising-like behavior to a distinctly separate universality class where the coupling between  $T_s$ and  $T_N$  has been substantially weakened. Whereas the previously observed coupling between the primary structural and magnetic order parameters in the undoped 122 systems renders identical critical exponents for both phase transitions, surprisingly, we find that far from the critical point (where  $T_N < T_S$ ) both critical exponents modeling the structural and magnetic phase behaviors in the iron arsenides change in an identical fashion. Thus, even for systems such as LaFeAsO, where  $T_s = 1.1T_N$ , the structural, orthorhombic distortion develops in a manner identical to the onset of AF order in the material. These empirical observations of potentially universal magnetic and structural behaviors among the various classes of iron pnictide high  $T_c$ 's hold promising implications for the identification of a common role for magnetism within their superconducting properties.

# II. MAGNETISM IN K2NiF4 AND BaFe2As2

Our previous investigation of AF order in the BaFe<sub>2</sub>As<sub>2</sub> (Ba-122) compound<sup>18</sup> revealed that the magnetic order parameter was well modeled by a simple power-law behavior of  $(1 - \frac{T}{T_N})^{\beta}$  with an exponent of  $\beta$ =0.125 thereby suggesting that the universality class of the AF phase transition was the same as that of the 2D-Ising model. While a weakly first-order component to the phase transition could not be entirely precluded, our previous analysis placed a limit on any possible linear, first-order trade off between phases to less than 0.5 K of  $T_N$ . The remarkable range in reduced temperature over which the simple power-law models the magnetic phase transition in BaFe<sub>2</sub>As<sub>2</sub> is reminiscent of the phase behavior



FIG. 2. (Color online) Comparison plots of the magnetic order parameters in BaFe<sub>2</sub>As<sub>2</sub> and K<sub>2</sub>NiF<sub>4</sub> with normalization described in the text. The solid yellow line denotes the expected behavior for the ideal 2D-Ising system. (a) Neutron measurements of  $M(\frac{T}{T_N})$  for BaFe<sub>2</sub>As<sub>2</sub> and K<sub>2</sub>NiF<sub>4</sub> taken from Refs. 18 and 22, respectively. (b) Plot showing  $M^2(\frac{T}{T_N})$  for BaFe<sub>2</sub>As<sub>2</sub> fit with a power-law model convolved with a Gaussian distribution of  $T_N$ 's as described in the text.

in known low-dimensional magnets such as the prototypical 2D magnet  $K_2NiF_4$  (Ref. 22) which stands as a useful standard for comparison when examining the phase-transition behavior in BaFe<sub>2</sub>As<sub>2</sub>.

 $K_2NiF_4$  is a model 2D system with a crossover from 2D Heisenberg behavior at low temperatures to anisotropydriven 2D Ising behavior in the vicinity of  $T_N$ . Figure 2(a) displays a comparison of the magnetic order parameters, M, as a function of  $(1 - T/T_N)$  for both systems K<sub>2</sub>NiF<sub>4</sub> and  $BaFe_2As_2$  where data for  $BaFe_2As_2$  are taken from Ref. 18 and the data for  $K_2NiF_4$  are extracted from Ref. 22. In order to facilitate a comparison of the phase-transition behavior in each respective system, all data were normalized at [1  $-(T/T_N)$ ]=0.1, where  $\approx 75\%$  of the saturated moment had been reached. Examination of this comparison highlights the striking similarity between the evolution of the magnetic order parameters in Ba-122 and K<sub>2</sub>NiF<sub>4</sub> where the critical behaviors of both systems with freely refined exponents of  $\beta$ =0.10 and  $\beta$ =0.14, respectively, bracket the ideal 2D Ising curve with  $\beta$ =0.125. In particular, this demonstrates explicitly that the sharp onset of antiferromagnetism in BaFe<sub>2</sub>As<sub>2</sub> mirrors that of a known second-order 2D-Heisenberg-Ising AF phase transition and thus that it cannot be easily dismissed as an artifact from an obscured first-order phase transition.

At temperatures immediately above  $T_N$ , appreciably more magnetic scattering at the three-dimensional (3D) ordering wave vector persists in BaFe<sub>2</sub>As<sub>2</sub> relative to K<sub>2</sub>NiF<sub>4</sub>. The origin of this tail of scattering above  $T_N$  is likely due to a small distribution of AF ordering temperatures throughout the bulk of the sample. This can be confirmed by fitting the expected power-law behavior of the order parameter weighted by a Gaussian distribution of  $T_N$ 's within the sample. The result of such a fit to the form  $M(T)^2 = B^2 \int (1 + M(T)^2) dt$  $-\frac{T}{T_{N}}^{2} \beta e^{-(T-\langle T_{N} \rangle)/2\sigma^{2}}$  is shown in Fig. 2(b) and the fit gives values of  $\langle T \rangle = 136.12 \pm 0.03$  K,  $\beta = 0.106 \pm 0.002$ , and  $\sigma$ =0.28  $\pm$  0.02 K. The power law was forced to 0 for  $T > T_N$ and only data in the range 0.85 K  $< \frac{T}{T_N} < 1.025$  K were included. The excellent agreement between this Gaussianbroadened power-law description and the magnetic order parameter in BaFe<sub>2</sub>As<sub>2</sub> clearly demonstrates that the phase transition in its entirety can be effectively modeled by a simple power law with a  $\beta \approx 0.125$  and a slightly broadened  $T_N$  with full width at half maximum, FWHM=0.0048 in reduced temperature. Additionally, our subsequent fitting of the same data to the first-order form  $M^2(T) = [A_s + B(1 - \frac{T}{T_s})^{\beta}]^2$ yielded negligible  $A_s$  values and identical  $\beta$  values to the previous  $A_s \equiv 0$  fits, thus further supporting our claim of continuous behavior in BaFe<sub>2</sub>As<sub>2</sub>.

## III. MAGNETIC AND STRUCTURAL ORDER PARAMETERS IN THE IRON ARSENIDES

Having established that the magnetic phase transition in Ba-122 mirrors that of a known 2D-Heisenberg-Ising magnet, it is informative now to compare the magnetic order parameters between different classes of iron arsenides. In Fig. 3(a), measurements of magnetic order parameters squared are plotted for both Ba-122 and LaFeAsO, where an immediate difference between the temperature dependencies of the order parameters is apparent. The data for LaFeAsO are taken from Ref. 23 which also reports a clear commonality among the measured magnetic order parameters of the REFeAsO systems. Therefore from Fig. 3(a) it is clear that there is a fundamental difference in the phase-transition behavior of the Ba-122 and the 1111 class of iron arsenides where the order parameter in LaFeAsO fit between  $0.6 < \frac{T}{T_N}$ <1.05 to a simple power law gives a  $\beta = 0.22 \pm 0.06$ . The yellow dashed line in Fig. 3(a) shows a simple power-law function with a fixed  $\beta$ =0.25 overplotted with the LaFeAsO data. Unfortunately, due to the lack of detailed data close to the phase transition in LaFeAsO, the critical exponent for the magnetic order parameter cannot be determined precisely; however from Fig. 3(a) it is apparent that a simple  $\beta = 0.25$ provides a reasonable approximation to the magnetic phase behavior in this system. This renders a critical exponent  $\beta$  in the monolayer iron arsenides roughly two times larger than the  $\beta = 0.125$  reported to model the phase behavior in Ba-122.<sup>18</sup> Given that both systems possess the same intrinsic spin structures and magnetic ions, this sharp distinction is surprising and, we believe, of fundamental importance.

In noting that the strongly coupled structural and magnetic order parameters in Ba-122 are known to show an iden-



FIG. 3. (Color online) (a) Plot showing the comparison of  $M^2(\frac{T}{T_N})$  for BaFe<sub>2</sub>As<sub>2</sub> and LaFeAsO. The solid black line denotes the Gaussian convolved power-law fit from Fig. 2(b) and the dashed line shows the results of a power-law fit with  $\beta$ =0.25 as described in the text. (b) Overplot of the magnetic order parameter (data taken from Ref. 23) and the structural order parameter (data taken from Ref. 24) for the LaFeAsO system. The black and yellow lines are the results of the fits with  $\beta$  fixed to be 0.22 and 0.25, respectively.

tical temperature evolution,<sup>18</sup> it is clearly important to examine how the structural phase transition is modified in LaFeAsO where  $T_S$  and  $T_N$  are largely decoupled. An overlay of both the magnetic order-parameter data again taken from Ref. 23 and the structural order-parameter data extracted from Ref. 24 is plotted in Fig. 3(b). From this figure, it is immediately evident that, despite the splitting of  $T_{\rm S}$  and  $T_{\rm N}$ by over more than 14 K, the phase behaviors of both the magnetic and structural order parameters in LaFeAsO track one another quite precisely. The yellow solid line in Fig. 3(b) shows the results of a power-law fit with  $\beta = 0.25$  overlaid with both sets of data while the black line shows the results from the freely refined fit of the magnetic order parameter in LaFeAsO (discussed previously). This simple, empirical, comparison suggests that the magnetostructural coupling observed in Ba-122 and other undoped, bilayer pnictides is also reflected in the monolayer pnictides where  $T_S$  and  $T_N$  are no longer coincident. The primary effect of tuning away from the  $T_N = T_S$  critical point appears to be a renormalization of the critical exponents modeling both the structural and magnetic order parameters which are altered in the same fashion. Despite the significant difference in  $\beta$  values between the 122 and 1111 compounds, the structural and magnetic phase behaviors mirror one another identically in both systems,



FIG. 4. (Color online) Plot showing the comparison of  $M^2(\frac{T}{T_N})$  for systems in the 122, 1111, and 111 classes of iron arsenide superconductors with the normalization procedure between different systems described in the text. The solid black line and dashed black line denote the ideal 2D-Ising and 3D-Ising values for  $M^2(\frac{T}{T_N})$ , respectively. The collapse onto two distinct curves for systems where  $T_N = T_S$  and for systems where  $T_N < T_S$  is shown.

thus strongly suggesting that this is a universal feature within the parent phases of the iron arsenides.

In considering the immediate contrasts in the magnetic phases of the 122 and 1111 compounds which may lead to the large differences in their magnetic and structural phasetransition behaviors, the primary distinction is that the strongly coupled, concomitant, magnetostructural phase transitions in the 122 series splits in temperature within the 1111 compounds thereby potentially altering the critical spin behavior. In order to investigate this further, we examined data from the literature reporting the magnetic order parameters in lightly Ni-doped and Co-doped Ba-122 systems.7,19,25 Within these initial reports, it was observed that doping small amounts of electrons splits the magnetic and structural phase transitions in Ba-122, and from the reported data it can be clearly seen that the observed order parameter is dramatically modified from the parent system's behavior. In both cases, a simple power law with an exponent of  $\beta \approx 0.3$  was reported to model the magnetic phase transitions close to  $T_N$ .<sup>7,19</sup> In order to provide a more explicit comparison between these various systems, Fig. 4 overplots the magnetic order parameters of both of these doped Ba-122 systems along with the magnetic order parameters of several different iron arsenide parent systems extracted from the literature.7,9,18,19,25,26 Systems whose phase transitions are closely modeled by a  $\beta$ =0.125 were normalized together at  $T/T_N = 0.9 \approx 0.56 M^2 (\frac{T}{T_N})$ =0) and systems whose phase transitions were approximated by  $\beta \approx 0.25$  were normalized together at  $T/T_N = 0.68 \approx 0.56 M^2 (\frac{T}{T_N} = 0)$ . For clarity, the order parameters of each class of systems were set apart by cross normalizing the average values of the order parameters of the  $\beta \approx 0.25$  class and the  $\beta$ =0.125 class at 75% of their saturated values. In determining which data to include in this comparison, we attempted to select the highest quality data in the literature using single-crystal measurements when possible. As Sn-flux-grown crystals exhibit Sn incorporation which strongly renormalizes magnetic properties in certain 122 pnictides,<sup>27</sup> we utilized data only from studies performed on crystals grown via FeAs or self-flux methods.

Looking at Fig. 4, it is immediately clear that there are two distinct classes of transitions among the data plotted. Both 122 parent systems, SrFe<sub>2</sub>As<sub>2</sub> and BaFe<sub>2</sub>As<sub>2</sub>, collapse onto a single curve that is modeled well by the  $\beta = 0.125$ 2D-Ising order parameter. Data from doped 122 systems and from 1111 and 111 iron arsenide compounds in Fig. 4 reveal that, in systems where the structural and magnetic phase transitions separate in temperature, the resulting magnetic phase transition collapses onto a separate universal curve whose critical exponent is roughly double that of the  $T_N$  $=T_S$  materials. Within this second class of magnetic phase transitions, fits to the data between  $0.6 \le \frac{T}{T_{\rm M}} \le 1.1$  led to an average exponent of  $\beta = 0.27 \pm 0.03$  demonstrating that, once the structural phase transition decouples from the onset of magnetic order, the universality class of the magnetic phase transition is clearly altered. Currently however, the quality of data from these systems with broader, decoupled magnetic phase transitions precludes the determination of their precise critical exponents; however we have fit to the data in Fig. 4 one possibility of a crossover to a 3D-Ising order parameter with  $\beta = 0.325$ . A 3D-Heisenberg transition with  $\beta = 0.365$ (not shown) may also fall within the error of the data suggesting in both cases an increased dimensionality to the magnetic phase transition once decoupled from  $T_s$ .

#### **IV. DISCUSSION**

Several theories have been put forth examining the relationship between the higher temperature structural transition and the lower temperature magnetic phase transition in the 1111 series of pnicides. Work by Xu et al.28 considered the presence of a finite temperature 2D Ising order which can be envisioned as an electronic, nematic phase that forms prior to the onset of the spin-density-wave (SDW) order. Additionally, recent work by Chen et al.<sup>29</sup> has considered the presence of an orbital ordering parameter also possessing 2D-Ising symmetry stemming from a simple spin-orbital Hamiltonian. SDW order in these models freezes out due to the finite  $J_c$  coupling and is simply a reflection of the eventual onset of three-dimensional ordering within the system. However, to the best of our knowledge, no theoretical work exploring the expected spin behavior at the effective critical point in the 122 pnictides where both phase transitions are coincident in temperature has been performed.

The universality demonstrated in both the magnetic and structural order parameters among the various classes of iron pnictides, and specifically the peculiar coupling between the magnetic and structural phase transitions is suggestive of a universal symmetry breaking responsible for the origin of both primary order parameters. One potential model addressing this phenomenon has been presented by Cvetkovic and Tesanovic<sup>30</sup> via a valley-density-wave (VDW) approach. Since the generic VDW state in a fully symmetric approach represents one primary instability comprised of combined spin, charge, and orbital density waves derived from quasiparticle excitations across the Fermi surface (FS), the identical phase behavior of both the structural and magnetic phase transitions observed in the pnictides—even when offset by large ranges in onset temperatures—may lend itself to such a universal origin. This also further highlights that the identical evolution of the structural and magnetic order parameters in the 122 parent systems implies that the structural phase transition is not a simple, secondary, consequence of magnetic ordering coupling to the lattice through strain.

The effect of the drastic change in the in-plane exchange couplings of the 122 parent systems as the structural and magnetic phase transitions evolve must also be taken into account. From early inelastic neutron-scattering measurements on one such parent system, CaFe<sub>2</sub>As<sub>2</sub><sup>21</sup> it was reported that at low temperatures, within the orthorhombic phase, the magnetic exchange energies within the Fe planes conform to a highly asymmetric  $J_{1A}$ - $J_{1B}$ - $J_2$  Heisenberg model. While the in-plane exchange along the elongated a axis was reported to exhibit a strong AF coupling of  $J_{1A}$ =49.9 meV, the orthogonal exchange pathway along the baxis was observed to be  $J_{1B}$ =-5.7 meV. For the hightemperature tetragonal phase however, symmetry dictates that both of these exchange couplings be identical. An extraordinarily sharp shift in the  $J_1$  parameters upon cooling through the combined magnetic and structural phase transitions is therefore implied in these systems. Even if one assumes that the reported low-temperature  $J_{1A}$  and  $J_{1B}$  values are symmetrically split from their nominal high-temperature value, this drastic shift traverses an energy range of  $\approx$  319 K in the  $J_{1B}$  parameter in likely under the  $\approx 1$  K it takes for the structural distortion to reach 50% of its saturation value. Although the fitting error range reported by Zhao et al.<sup>21</sup> precludes any definitive statement regarding any dimensional crossover in this regime, the overall negative value of  $J_{1B}$ suggests a point during the magnetic phase transition in which the nearest-neighbor magnetic exchange along the baxis vanishes. Therefore, neglecting next-nearest-neighbor interactions, there potentially exists a narrow window during the onset of AF order in which the 122 parent systems are quasi-two dimensional in the [H, 0, L] plane. This drastic renormalization of the exchange parameters undergone during the course of the magnetic phase transition unfortunately renders the magnetic phase behavior in the 122 parent systems difficult to model. Detailed measurements of the magnetic exchange couplings in these materials, as they traverse from the tetragonal to orthorhombic structural symmetries, are an interesting and fundamentally necessary future step in the study of magnetism in these parent materials.

Experimental work on BaFe<sub>2</sub>As<sub>2</sub> has reported a reduction in the anisotropy gap of the spin excitations upon Ni doping onto the Fe site, potentially indicating an enhanced two dimensionality induced upon carrier doping.<sup>19</sup> This seems to run counter to our current observation of a transition from a regime in which the magnetic phase-transition behavior is consistent with that of a two-dimensional magnetic system in the BaFe<sub>2</sub>As<sub>2</sub> system to a regime in doped samples whose larger critical exponents (i.e.,  $\beta \approx 0.27$ ) suggest a new universality class with a more three-dimensional character. Photoemission experiments, however, have shown an abrupt change in the dimensionality of Co-doped Ba-122 where the two-dimensional electronic states of the parent Ba-122 system are tuned toward a more three-dimensional dispersion as Co is doped into the system.<sup>31,32</sup> Separate neutron studies of BaFe<sub>2</sub>As<sub>2</sub> have also reported largely two-dimensional spin fluctuations above  $T_S$  (Ref. 17) consistent with the picture of a two-dimensional magnetic order parameter close to  $T_N$  and qualitatively in agreement with the largely two-dimensional Fermi surface reported in recent photoemission measurements on BaFe<sub>2</sub>As<sub>2</sub> (Refs. 31 and 32) and CaFe<sub>2</sub>As<sub>2</sub>.<sup>33</sup>

However, when considering bulk measurements of the FS via quantum oscillation experiments, this picture of a quasitwo-dimensional FS correlating to a more two-dimensional magnetic phase behavior in the 122 pnictides breaks down. In particular, quantum oscillation measurements report a small, fully three-dimensional FS in the parent 122 iron pnictides<sup>34,35</sup> which contrasts with the definitively more twodimensional electronic structures within the 1111 class of systems.<sup>36</sup> Quantum oscillation measurements of how the FS evolves upon suppressing  $T_N$  thus far remain confined to the study of weakly/nonsuperconducting, analog materials such as LaFePO (Ref. 36) and SrFe<sub>2</sub>P<sub>2</sub>.<sup>37</sup> While this precludes any direct analysis of how the bulk FS evolves relative to the magnetic phase behavior, the picture of the FS derived from these quantum oscillation measurements suggests that the universal features of the magnetic phase behavior in the iron pnictides are seemingly decoupled from the topological details of the FS. The only relevant parameter which seemingly controls the magnetic and structural phase behaviors in the iron arsenides is the proximity of  $T_S$  and  $T_N$ . While the relative proximities of the onsets of the structural and magnetic phase transitions are likely indirectly coupled to the effective dimensionality of the system, they do not appear to follow the general trends in the bulk FS dimensionality in an obvious way. This observation may suggest a larger role for local-moment physics in the formation of the AF ordered states in the pnictides than a simple spin-density-wave nesting picture. To date, however, there has been no comprehensive experimental investigation into the detailed evolution of the dimensionality of spin fluctuations in a 122 system as it is tuned from the quasi-2D high-temperature regime above  $T_S$  to the anisotropic, three-dimensional magnetic and electronic phases far below  $T_N$ . Future work exploring this along with further theoretical efforts detailing the coupled phase transitions in the pnictides are needed in order to understand the seemingly universal structural and magnetic phase behaviors within these materials.

#### **V. CONCLUSION**

In summary, we have presented an empirical analysis of the magnetic order parameters in the primary classes of the iron arsenide superconductors. Within the undoped, bilayer 122 class of iron pnictides where  $T_N=T_S$ , both the magnetic and structural phase transitions are well modeled by a 2D-Ising order parameter; however, upon doping, the magnetic and structural phase transitions alter to exhibit a critical  $\beta$  $\approx 0.27$ . The likely cause of this crossover in the phase behavior is the decoupling of the magnetic and structural transitions where alternate classes of iron pnictides with  $T_N$  $< T_{\rm S}$  collapse onto the same seemingly universal curve. Two distinct behaviors therefore appear in the magnetic and structural phase transitions of the iron arsenides: The first of these occurs when the structural and magnetic phase transitions coincide at a multicritical point and the second emerges once these two transitions no longer coincide rendering strongly renormalized critical exponents for both. The critical behavior of the iron pnictides, therefore, seems to transition from one which parallels a known 2D magnet when  $T_N = T_S$  to one consistent with a more three-dimensional character upon tuning away from the critical point. This observation currently contradicts several existing arguments for more twodimensional behavior reported in the Ba-122 system upon introducing charge carriers, thus suggesting that new theoretical insight is needed.

Additionally, a better experimental picture is needed regarding the complex modification in spin behavior under-

- <sup>1</sup>M. A. Kastner, R. J. Birgeneau, G. Shirane, and Y. Endoh, Rev. Mod. Phys. **70**, 897 (1998).
- <sup>2</sup>Q. Huang, J. Zhao, J. W. Lynn, G. F. Chen, J. L. Luo, N. L. Wang, and P. Dai, Phys. Rev. B **78**, 054529 (2008).
- <sup>3</sup>J.-H. Chu, J. G. Analytis, C. Kucharczyk, and I. R. Fisher, Phys. Rev. B **79**, 014506 (2009).
- <sup>4</sup>C. de la Cruz, Q. Huang, J. W. Lynn, J. Li, W. Ratcliff II, J. L. Zarestky, H. A. Mook, G. F. Chen, J. L. Luo, N. L. Wang, and P. Dai, Nature (London) **453**, 899 (2008).
- <sup>5</sup>H. Luetkens, H.-H. Klauss, M. Kraken, F. J. Litterst, T. Dellmann, R. Klingeler, C. Hess, R. Khasanov, A. Amato, C. Baines, M. Kosmala, O. J. Schumann, M. Braden, J. Hamann-Borrero, N. Leps, A. Kondrat, G. Behr, J. Werner, and B. Büchner, Nature Mater. **8**, 305 (2009).
- <sup>6</sup>J. Zhao, Q. Huang, C. de la Cruz, S. Li, J. W. Lynn, M. A. Green, Y. Chen, G. F. Chen, G. Li, Z. Li, J. L. Luo, N. L. Wang, and P. Dai, Nature Mater. 7, 953 (2008).
- <sup>7</sup>D. K. Pratt, W. Tian, A. Kreyssig, J. L. Zarestky, S. Nandi, N. Ni, S. L. Bud'ko, P. C. Canfield, A. I. Goldman, and R. J. Mc-Queeney, Phys. Rev. Lett. **103**, 087001 (2009).
- <sup>8</sup>G. Shirane, Y. Endoh, R. Birgeneau, M. Kastner, Y. Hidaka, M. Oda, M. Suzuki, and T. Murakami, Phys. Rev. Lett. **59**, 1613 (1987).
- <sup>9</sup>S. Li, C. de la Cruz, Q. Huang, G. F. Chen, T.-L. Xia, J. L. Luo, N. L. Wang, and P. Dai, Phys. Rev. B **80**, 020504(R) (2009).
- <sup>10</sup>Q. Huang, Y. Qiu, W. Bao, M. A. Green, J. W. Lynn, Y. C. Gasparovic, T. Wu, G. Wu, and X. H. Chen, Phys. Rev. Lett. **101**, 257003 (2008).
- <sup>11</sup>M. Kofu, Y. Qiu, W. Bao, S.-H. Lee, S. Chang, T. Wu, G. Wu, and X. H. Chen, New J. Phys. **11**, 055001 (2009).
- <sup>12</sup>C. Krellner, N. Caroca-Canales, A. Jesche, H. Rosner, A. Ormeci, and C. Geibel, Phys. Rev. B 78, 100504(R) (2008).
- <sup>13</sup>A. I. Goldman, D. N. Argyriou, B. Ouladdiaf, T. Chatterji, A. Kreyssig, S. Nandi, N. Ni, S. L. Bud'ko, P. C. Canfield, and R.

gone during the magnetic phase transition in the parent 122 iron pnictides. The evolution of the in-plane exchange couplings as these systems are cooled into their orthorhombic phases must be fully elucidated prior to any microscopic understanding of the 2D-Ising-like phase behavior of these materials. Our analysis demonstrates that further theoretical and experimental work exploring the phase behavior in the critical magnetism of the iron pnictides have the possibility of revealing exciting new physics relevant to the fundamental symmetries in the magnetism of these compounds.

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J. McQueeney, Phys. Rev. B 78, 100506(R) (2008).

- <sup>14</sup>K. Kitagawa, N. Katayama, K. Ohgushi, M. Yoshida, and M. Takigawa, J. Phys. Soc. Jpn. 77, 114709 (2008).
- <sup>15</sup>M. Tegel, M. Rotter, V. Weiß, F. M. Schappacher, R. Pöttgen, and D. Johrendt, J. Phys.: Condens. Matter **20**, 452201 (2008).
- <sup>16</sup>M. Rotter, M. Tegel, D. Johrendt, I. Schellenberg, W. Hermes, and R. Pöttgen, Phys. Rev. B 78, 020503(R) (2008).
- <sup>17</sup>K. Matan, R. Morinaga, K. Iida, and T. J. Sato, Phys. Rev. B **79**, 054526 (2009).
- <sup>18</sup>S. D. Wilson, Z. Yamani, C. R. Rotundu, B. Freelon, E. Bourret-Courchesne, and R. J. Birgeneau, Phys. Rev. B **79**, 184519 (2009).
- <sup>19</sup>L. W. Harriger, A. Schneidewind, S. Li, J. Zhao, Z. Li, W. Lu, X. Dong, F. Zhou, Z. Zhao, J. Hu, and P. Dai, Phys. Rev. Lett. **103**, 087005 (2009).
- <sup>20</sup>C. Lester, J.-H. Chu, J. G. Analytis, S. C. Capelli, A. S. Erickson, C. L. Condron, M. F. Toney, I. R. Fisher, and S. M. Hayden, Phys. Rev. B **79**, 144523 (2009).
- <sup>21</sup>J. Zhao, D. T. Adroja, D.-X. Yao, R. Bewley, S. Li, X. F. Wang, G. Wu, X. H. Chen, J. Hu, and P. Dai, Nat. Phys. **5**, 555 (2009).
- <sup>22</sup>R. J. Birgeneau, H. J. Guggenheim, and G. Shirane, Phys. Rev. B 1, 2211 (1970).
- <sup>23</sup>H. Maeter, H. Luetkens, Y. G. Pashkevich, A. Kwadrin, R. Khasanov, A. Amato, A. A. Gusev, K. V. Lamonova, D. A. Chervinskii, R. Klingeler, C. Hess, G. Behr, B. Büchner, and H.-H. Klauss, Phys. Rev. B **80**, 094524 (2009).
- <sup>24</sup>J.-Q. Yan, S. Nandi, J. L. Zarestky, W. Tian, A. Kreyssig, B. Jensen, A. Kracher, K. W. Dennis, R. J. McQueeney, A. I. Goldman *et al.*, arXiv:0909.3510 (unpublished).
- <sup>25</sup> A. D. Christianson, M. D. Lumsden, S. E. Nagler, G. J. Mac-Dougall, M. A. McGuire, A. S. Sefat, R. Jin, B. C. Sales, and D. Mandrus, Phys. Rev. Lett. **103**, 087002 (2009).
- <sup>26</sup>J. Zhao, W. Ratcliff, J. W. Lynn, G. F. Chen, J. L. Luo, N. L. Wang, J. Hu, and P. Dai, Phys. Rev. B **78**, 140504(R) (2008).

- <sup>27</sup>Y. Su, P. Link, A. Schneidewind, T. Wolf, P. Adelmann, Y. Xiao, M. Meven, R. Mittal, M. Rotter, D. Johrendt, Th. Brueckel, and M. Loewenhaupt, Phys. Rev. B **79**, 064504 (2009).
- <sup>28</sup>G. Xu, W. Ming, Y. Yao, X. Dai, S. C. Zhang, and Z. Fang, EPL 82, 67002 (2008).
- <sup>29</sup>C.-C. Chen, B. Moritz, J. van den Brink, T. P. Devereaux, and R. R. P. Singh, Phys. Rev. B **80**, 180418(R) (2009).
- <sup>30</sup>V. Cvetkovic and Z. Tesanovic, Phys. Rev. B **80**, 024512 (2009).
- <sup>31</sup>S. Thirupathaiah, S. de Jong, R. Ovsyannikov, H. Dürr, A. Varykhalov, R. Follath, Y. Huang, R. Huisman, M. Golden, Y.-Z. Zhang *et al.*, arXiv:0910.0147 (unpublished).
- <sup>32</sup>P. Vilmercati, A. Fedorov, I. Vobornik, U. Manju, G. Panaccione, A. Goldoni, A. S. Sefat, M. A. McGuire, B. C. Sales, R. Jin *et al.*, Phys. Rev. B **79**, 220503(R) (2009).
- <sup>33</sup>C. Liu, T. Kondo, N. Ni, A. D. Palczewski, A. Bostwick, G. D.

Samolyuk, R. Khasanov, M. Shi, E. Rotenberg, S. L. Budko, P. C. Canfield, and A. Kaminski, Phys. Rev. Lett. **102**, 167004 (2009).

- <sup>34</sup>J. G. Analytis, R. D. McDonald, J.-H. Chu, S. C. Riggs, A. F. Bangura, C. Kucharczyk, M. Johannes, and I. R. Fisher, Phys. Rev. B 80, 064507 (2009).
- <sup>35</sup>S. E. Sebastian, J. Gillett, N. Harrison, P. H. C. Lau, D. J. Singh, C. H. Mielke, and G. G. Lonzarich, J. Phys.: Condens. Matter 20, 422203 (2008).
- <sup>36</sup>A. I. Coldea, J. D. Fletcher, A. Carrington, J. G. Analytis, A. F. Bangura, J.-H. Chu, A. S. Erickson, I. R. Fisher, N. E. Hussey, and R. D. McDonald, Phys. Rev. Lett. **101**, 216402 (2008).
- <sup>37</sup>J. G. Analytis, C. M. J. Andrew, A. I. Coldea, A. McCollam, J.-H. Chu, R. D. McDonald, I. R. Fisher, and A. Carrington, Phys. Rev. Lett. **103**, 076401 (2009).