Critical behavior of the spin density wave transition in underdoped Ba(Fe1−xCox)2As2 (x ≤ 0.05): 75As NMR investigation

F. L. Ning,1,4 M. Fu,2 D. A. Torchetti,2 T. Imai,2,3 A. S. Sefat,4 P. Cheng,5 B. Shen,5 and H.-H. Wen3,5,6

1Department of Physics, Zhejiang University, Hangzhou 310027, P. R. China
2Department of Physics and Astronomy, McMaster University, Hamilton, Ontario, Canada L8S4M1
3Canadian Institute for Advanced Research, Toronto, Ontario, Canada MSG1Z8
4Materials Science and Technology Division, Oak Ridge National Laboratory, Tennessee 37831, USA
5National Laboratory for Superconductivity, Institute of Physics and Beijing National Laboratory for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100190, P. R. China
6Center for Superconducting Physics and Materials, National Laboratory for Solid State Microstructures, Department of Physics, Nanjing University, Nanjing 210093, P. R. China

(Received 4 April 2014; revised manuscript received 20 May 2014; published 25 June 2014)

We investigate the nature of the SDW (spin density wave) transition in the underdoped regime of an iron-based high-Τc superconductor Ba(Fe1−xCox)2As2 by 75As NMR, with primary focus on a composition with x = 0.02 (ΤSDW = 99 K). We demonstrate that critical slowing down toward the three-dimensional SDW transition sets in at the tetragonal to orthorhombic structural phase transition Τt = 105 K, suggesting strong interplay between structural distortion and spin correlations. In the critical regime between Τt and ΤSDW, the dynamical structure factor of electron spins S(q, 0ω) measured with the longitudinal NMR relaxation rate 1/Τ1 exhibits a divergent behavior obeying a power law 1/Τ1 ∝ ΣqS(q, 0ω) ∼ (Τ/ΤSDW − 1)−δ with the critical exponent δ ∼ 0.33.

DOI: 10.1103/PhysRevB.89.214511 PACS number(s): 74.70.Xa, 76.60.–k

I. INTRODUCTION

The discovery of superconductivity with Τc as high as 28 ~ 55 K in iron pnictides [1–5] has regenerated strong interest in the research of high-temperature superconductivity. The parent compound of the so-called 122 ferropnictides, BaFe2As2, is a semimetallic antiferromagnet; upon cooling, BaFe2As2 undergoes a first-order spin density wave (SDW) transition at ΤSDW ∼ 135 K, accompanied by a tetragonal to orthorhombic structural phase transition at Τt (= ΤSDW) [4,6–8]. Doping a few percent of Co into the Fe sites of BaFe2As2 quickly suppresses ΤSDW [9,10] as well as Τt [11], as summarized in Fig. 1. In the lightly Co-doped regime, the structural phase transition takes place first upon cooling, followed by the SDW transition in the orthorhombic phase [11,12].

Superconductivity with optimized Τc ∼ 25 K appears when Τt and ΤSDW are completely suppressed by 6% ~ 8% Co doping [5,9–14]. The nature and origin of the SDW ordering, and its potential relation to the superconducting mechanism, are the subject of intense debates [15].

In this work, we investigate the critical behavior of the SDW transition and its interplay with the structural transition in lightly Co-doped single crystals of Ba(Fe1−xCox)2As2 with x = 0.02, 0.04, and 0.05 based on 75As NMR measurements. We will place our primary focus on a composition with x = 0.02; thanks to their relatively sharp NMR lines, experimental characterizations of structural and SDW phase transitions are straightforward for this composition. We demonstrate that the structural transition at Τt = 105 K triggers the critical slowing down of spin dynamics toward the three-dimensional SDW transition at ΤSDW = 99 K. We found that the critical exponent for the divergence of the dynamical structure factor of electron spins S(q, 0ω) near the SDW transition is different from δ = 1/2 often attributed to itinerant electron magnetism, such as metallic Cr [16]. Instead, we found δ ∼ 0.33. This value is nearly identical with the case of a Mott insulator CuO with δ = 0.33 ± 0.01 [17], and is in reasonable agreement with the theoretically predicted value of δ = v/2 ~ 0.35 for insulating three-dimensional (3D) Heisenberg antiferromagnets [18–21]. Here, v ~ 0.7 is the critical exponent for the spin-spin correlation length ξ, and ξ ∼ (Τ/ΤSDW − 1)−v. We also demonstrate that Co doping enhances the density of states D(EF) of the reconstructed Fermi surfaces below ΤSDW roughly in proportion to x, based on the enhancement of 1/Τ1Τ at low temperatures.

The rest of this paper is organized as follows. In Sec. II, we will briefly describe experimental procedures. In Sec. III, we will discuss our results in the paramagnetic state above ΤSDW, followed by brief discussions about the SDW ordered state. We will conclude in Sec. IV.

II. EXPERIMENTAL METHODS

We grew single crystals of Ba(Fe1−xCox)2As2 from FeAs flux [5,14]. We carried out NMR measurements using the standard pulsed NMR techniques. For x = 0.02, we cleaved a small piece of shiny crystal from a much larger boule used for our previous report [22]. The total mass of the smaller crystal used for this work is about ~7 mg. It was necessary to use the smaller piece to ensure high homogeneity of the sample. In fact, we found no evidence for a stretched recovery of Τ1 [23] in our small homogeneous crystal of x = 0.02, contrary to an earlier report that an x = 0.022 crystal [24] and lightly doped LaFeAsO1−xF6 crystals [25] exhibit a large distribution of Τ1, which implies a large distribution of ΤSDW. From the sharpness of the divergent behavior of 1/Τ1 and the NMR linewidth, we estimate the upper bound of the distribution of Τt and ΤSDW as little as ~0.5 K in our small x = 0.02 crystal. Moreover,
we could resolve the fine structures of the NMR line shapes in the magnetically ordered state below \( T_{SDW} \) [see Figs. 2(b) and 2(c)], which we were unable to detect in our earlier study using a larger, inhomogeneous crystal [22]. Due to the poor signal-to-noise ratio arising from the small volume of the crystal and long relaxation time \( T_1 \), the NMR data acquisition is extremely time consuming below \( T_{SDW} \); it took up to \( \sim 10 \) days of continuous signal averaging to complete one set of NMR line-shape measurements at a given temperature.

Small single-crystal samples used for other compositions with \( x = 0.04 \) and 0.05 are identical with those used in our previous studies [10,26]. We found stretched forms of \( T_1 \) recovery only for \( x = 0.05 \) below \( \sim 70 \) K, analogous to the earlier report [24]. It is worth recalling that Co substitution is known to suppress spin fluctuations locally at Co sites, as evidenced by temperature independent \( 1/T_1 \) observed at Co sites at low temperatures [27]. A level of distribution in the electronic properties in the alloyed samples of Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)As\(_2\) is therefore naturally expected, as we demonstrated earlier from the variation of \( 1/T_1 \) within a single NMR peak of a given composition [10]. But, none of the key findings and conclusions in this work rely on the \( x = 0.05 \) sample at low temperatures, and hence the issue of the inhomogeneity induced by Co substitution is beyond the scope of this work.

III. RESULTS AND DISCUSSIONS

A. \(^{75}\)As NMR line shape, width, and Knight shift

In Fig. 2(a), we present a representative field-swept \(^{75}\)As NMR line shape of Ba(Fe\(_{0.96}\)Co\(_{0.04}\))\(_2\)As\(_2\) observed at a fixed NMR frequency of \( \omega_n/2\pi = 43.503 \) MHz in the paramagnetic state above \( T_{SDW} \). In general, the nuclear spin Hamiltonian can be expressed as a summation of the Zeeman and nuclear quadrupole interaction terms

\[
H = -\gamma_n h \vec{B} \cdot \vec{I} + \frac{\hbar \nu_0}{6} \left( 3I_z^2 - I(I + 1) + \frac{1}{2} \eta(I_x^2 + I_y^2) \right),
\]

(1)
where the $^{75}\text{As}$ nuclear gyromagnetic ratio is $\gamma_n/2\pi = 7.2919 \text{ MHz/T}$, $h$ is Planck’s constant, and $\mathbf{I}$ represents the nuclear spin. Since $^{75}\text{As}$ has nuclear spin $I = \frac{3}{2}$, we observe three transitions from $I_z = \frac{3}{2}$, $\frac{1}{2}$, and $-\frac{1}{2}$ to $\frac{1}{2}$, $\frac{1}{2}$, and $-\frac{1}{2}$, respectively, in the NMR line shape: the sharp central peak arises from the $I_z = \frac{1}{2}$ to $\frac{1}{2}$ transition ($m = 0$); additionally, two broad satellite peaks arise from the $I_z = \pm \frac{1}{2}$ transitions ($m = \pm 1$), separated by $75v_Q^0$. The nuclear quadrupole interaction frequency $v_Q^0$ along the $c$ axis is proportional to the electric field gradient (EFG) at the observed $^{75}\text{As}$ site, and $\eta$ is the asymmetry parameter of the EFG, $\eta = |v_a^0 - v_b^0|/v_Q^0$. Due to the tetragonal symmetry at the $^{75}\text{As}$ sites, $\eta = 0$ above $T_s$. Co doping induces substantial disorder in the lattice, reflected on the distribution of $75v_Q^0).

$\mathbf{B}$ is the summation of the external field $\mathbf{B}_{\text{ext}}$ and the time-averaged hyperfine fields from nearby electron spins $\mathbf{B}_{\text{hf}}$, i.e., $\mathbf{B} = \mathbf{B}_{\text{ext}} + \mathbf{B}_{\text{hf}}$. In the paramagnetic state, the central peak frequency is slightly shifted (i.e., “Knight shift”) due to small hyperfine fields induced by polarized electron spins nearby. Since the spin polarization induced by $\mathbf{B}_{\text{ext}}$ is proportional to spin susceptibility $\chi_{\text{spin}}$, we can measure the latter by accurately determining the central peak position [28]. In the SDW ordered state, static $\mathbf{B}_{\text{hf}}$ induced by ordered magnetic moments in the vicinity of the observed $^{75}\text{As}$ nuclear spins dramatically affects the NMR line shapes, as shown in Figs. 2(b) and 2(c). We will come back to this point below in Sec. III C.

We summarize the temperature dependence of the paramagnetic NMR Knight shift $75K$ and the FWHM (full width at half maximum) of the central peak frequency in Figs. 3 and 4, respectively. To ensure high accuracy, we conducted these measurements by taking the FFT (fast Fourier transform) of the spin-echo envelope in a fixed magnetic field. The NMR Knight shift $75K = A_{\text{hf}}\chi_{\text{spin}} + K_{\text{chem}}$ probes the local spin susceptibility $\chi_{\text{spin}}$ via hyperfine coupling $A_{\text{hf}}$, $K_{\text{chem}} (\sim 0.2\%$ or less for $x = 0.02$) is a temperature-independent chemical shift [27]. Our new results of $75K$ in Ba(Fe$_{0.98}$Co$_{0.02}$)$_2$As$_2$ are analogous to those observed for other compositions [10,26,27]: $75K$ decreases with temperature, and tends to level off near $\sim 100\ K$ [29]. See [26] for detailed analysis of $75K$ based on fitting the data with a pseudogap $\Delta_{PC}/k_B \sim 450\ K$.

One interesting aspect of Fig. 3 is that $75K$ exhibits a noticeable drop below $105.0 \pm 0.5\ K$ for Ba(Fe$_{0.98}$Co$_{0.02}$)$_2$As$_2$. This anomaly is accompanied by a sudden onset of the divergent behavior of FWHM, as shown in Fig. 3. We note that FWHM indeed diverges below $T_{\text{SDW}} = 99.0 \pm 0.5\ K$, where the emergence of static hyperfine magnetic field $\mathbf{B}_{\text{hf}}$ splits the NMR line in the SDW ordered state, as shown in Figs. 2(b) and 2(c). We found analogous anomalies of $75K$ and FWHM for Co 4% and 5% doped samples at $77 \pm 2\ K$ and $55 \pm 2\ K$, respectively, as shown in Figs. 3 and 4. We summarize the concentration dependence of these anomalies in Fig. 1. Clearly, these anomalies are related to the structural phase transition at $T_s$ [11,12].

Having identified the signature of the structural phase transition at $T_s$ in our NMR data for the central transition, we also searched for an anomaly in the nuclear quadrupole frequency $v_Q^0$ by measuring the splitting between the central and satellite peaks. We recall that, in typical second-order structural phase transitions such as the high-temperature tetragonal to low-temperature orthorhombic phase transition in the undoped and Sr-doped La$_2$CuO$_4$ high-$T_c$ cuprates, one could even observe a $\lambda$-like kink in the temperature dependence of $v_Q^0$ [30]. We summarize our results for Ba(Fe$_{0.98}$Co$_{0.02}$)$_2$As$_2$ in Fig. 5. (High-precision determination of $75v_Q^0$ is rather difficult for higher Co concentrations because the satellite peaks become very broad due to disorder [27].) In the case of undoped BaFe$_2$As$_2$, $75v_Q^0$ exhibits a step at the first-order structural
transition \( T_s = 135 \) K [8], but we find practically no anomaly at \( T_s = 105.0 \) K for the Co 2% sample. In general, when the lattice contracts with decreasing temperature, the lattice contribution to the electric field gradient (EFG), and hence to \( \nu_Q \), increases. Our finding that \( \nu_Q \) smoothly decreases with temperature might be an indication that there is a sizable onsite ionic contribution with an opposite sign.

It is not clear why \( \nu_Q \) does not exhibit a clear anomaly at \( T_s \) for the Co 2% doped sample. One possible scenario is that the influence of structural distortion on \( \nu_Q \) sites becomes so subtle under the presence of Co dopants that the change of \( \nu_Q \) also becomes extremely small. We also recall that softening of the lattice stiffness begins at unusually high temperatures in Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)As\(_2\), and has been speculated to be the consequence of antiferromagnetic correlations [31–33]. Perhaps the effects of orthorhombic distortion on \( \nu_Q \) appear progressively from much higher temperature than \( T_s \). In any event, the absence of a strong signature of structural anomaly in the temperature dependence of \( \nu_Q \) at \( T_s \) excludes the possibility that anomalies observed below \( T_s \) in Figs. 3 and 4 are a consequence of the subtle changes in the second-order quadrupole effects. In fact, we confirmed that the FWHM is approximately proportional to the magnitude of the applied magnetic field, hence the divergent behavior of FWHM below \( T_s \) is the consequence of magnetic effects.

Quite generally, divergence of the NMR linewidth precedes a magnetic phase transition through the divergence of dynamical spin susceptibility in the critical regime [28]. We also recall that the NMR Knight shift \( K \) reflects local paramagnetic spin susceptibility \( \chi_{\text{spin}} \); hence, the downturn in the temperature dependence of \( K \) below \( T_s \) is also consistent with suppression of \( \chi_{\text{spin}} \) due to strong antiferromagnetic short-range order. Thus, our findings in both Figs. 3 and 4 suggest that the structural phase transition at \( T_s \) drives the onset of strong 3D antiferromagnetic short-range order. This point is more vividly demonstrated through the divergent behavior of \( 1/T_1 \) in the next section.

**B. Critical spin dynamics near \( T_{\text{SDW}} \)**

In Fig. 6, we present \( \nu_Q \) nuclear spin-lattice relaxation rate \( 1/T_1 \) divided by temperature \( T \), i.e., \( 1/T_1 T \), observed for Ba(Fe\(_{0.98}\)Co\(_{0.02}\))\(_2\)As\(_2\). \( 1/T_1 T \) measures wave-vector \( q \) integral of the imaginary part of the dynamical electron spin susceptibility \( \chi'('q',\omega_k) \) weighted by the hyperfine form factor \( |A_{hy}('q')|^2 \) [34]. In the case of undoped BaFe\(_2\)As\(_2\), \( 1/T_1 T \) does not show divergent behavior at \( T_{\text{SDW}} \) expected for second-order magnetic phase transitions; instead, \( 1/T_1 T \) shows a step at 135 K because the SDW transition is first order [8]. In contrast, \( 1/T_1 T \) observed for Co 2% doped sample exhibits strongly divergent behavior near \( T_{\text{SDW}} \) in the geometry of \( B_{\text{ext}} \parallel ab \). In this configuration, \( 1/T_1 T \) probes fluctuations of hyperfine fields both along the \( c \) axis and \( ab \) plane. The divergent signature is less prominent for \( B_{\text{ext}} \parallel c \) because \( 1/T_1 T \) probes fluctuating hyperfine fields only within the \( ab \) plane, and the transferred hyperfine field \( A_{hy}('q') \) becomes vanishingly small for staggered wave vectors in this configuration [8,15,34]. In other words, it is advantageous to use the \( B_{\text{ext}} \parallel ab \) geometry to probe the critical behavior of the SDW transition.

Accordingly, in what follows, we focus our attention on \( 1/T_1 T \) measured in \( B_{\text{ext}} \parallel ab \). In Fig. 7, we show \( 1/T_1 T \) in a semi-log scale for various Co concentrations. To avoid confusion, we show only the results above \( T_{\text{SDW}} \) (for \( x \leq 5\% \)) or \( T_s \) (for \( x = 8\% \) and 12%) in the next section.

![FIG. 5. (Color online) The c-axis component of the 75As nuclear quadrupole frequency \( \nu_Q \) in (▲) Ba(Fe\(_{0.98}\)Co\(_{0.02}\))\(_2\)As\(_2\) (this work), and (■) undoped BaFe\(_2\)As\(_2\) [8]. Downward arrows mark \( T_s \), while vertical dotted line represents \( T_{\text{SDW}} \). We were unable to determine \( T_{\text{SDW}} \) accurately below \( T_{\text{SDW}} = 99.0 \) K except at 4.2 K due to extremely broad line profiles [see Fig. 2(b)].](image1)

![FIG. 6. (Color online) \( 1/T_1 T \) observed for Ba(Fe\(_{0.98}\)Co\(_{0.02}\))\(_2\)As\(_2\) under the external magnetic field \( B_{\text{ext}} \parallel ab \) (▲) or \( B_{\text{ext}} \parallel c \) (△). For comparison, we also show the results of BaFe\(_2\)As\(_2\) for \( B_{\text{ext}} \parallel ab \) (■) and \( B_{\text{ext}} \parallel c \) (□) [8]. Vertical dotted lines represent \( T_{\text{SDW}} \), while solid arrows mark \( T_s \). Solid curves are a Curie-Weiss fit (see main text). Notice that the Curie-Weiss fit breaks down at \( T_s = 105.0 \) K, and \( 1/T_1 T \) begins to blow up toward \( T_{\text{SDW}} = 99.0 \) K. Inset: the concentration \( x \) dependence of \( 1/T_1 T \) at 4.2 K for \( B_{\text{ext}} \parallel c \). The solid curve is a parabolic fit.](image2)
Curie-Weiss fit captures the temperature and concentration dependence of the susceptibility. As already discussed in detail in Ref. [26], the phenomenological Curie-Weiss fit incorporates a background term due to a pseudogap (see main text). The second, activation term in the fit represents the contributions of spin fluctuations toward the eventual three-dimensional SDW order. Analogous interplay between the spin and lattice degrees of freedom was also observed for LaFeAsO [40].

1/T_1 T = C/(T − θ) + A exp(−Δ_{PG}/k_B T) [26]. C and A are fitting parameters, and θ is the Weiss temperature of the staggered susceptibility χ_s(q,ω_n) near the ordering vector q. The concentration dependence of θ thus obtained is summarized in Fig. 1. Note that we have reversed the sign convention for θ in the present work (i.e., −θ in Fig. 1 corresponds to +θ in [26]). The second, activation term in the fit represents the background contributions which decrease with temperature, reflecting the pseudogap-like signature commonly observed for iron-pnictide and iron-selenide superconductors [26,35–39]. As already discussed in detail in [26], the phenomenological Curie-Weiss fit captures the temperature and concentration dependence of χ_s(q,ω_n) remarkably well, including the new results for the Co 2% doped sample. The Curie-Weiss behavior of 1/T_1 T reflects the fact that, upon cooling, short-range antiferromagnetic correlations slowly grow toward T_{SDW}. θ reverses its sign above the quantum critical point x_c ~ 0.065, which implies that Fe spins are not destined to order above x_c. Remarkably, the optimally superconducting composition with the maximum T_c ~ 25 K is located in the vicinity of x_c, hinting at the link between the superconducting mechanism and spin fluctuations [26].

Another important feature of Figs. 6 and 7 which we did not discuss explicitly in [26] is that the phenomenological Curie-Weiss fit breaks down below T_c. Extrapolation of the fit to below T_c underestimates the data points near the SDW phase transition for Co 2%, 4%, and 5%, and strong divergent behavior sets in at T_c. In other words, the three-dimensional short-range order sets in at the tetragonal to orthorhombic structural phase transition, which is prerequisite to the critical slowing down of spin fluctuations toward the eventual three-dimensional SDW order. Analogous interplay between the spin and lattice degrees of freedom was also observed for LaFeAsO [40].

In Fig. 8, we plot 1/T_1 of three underdoped compositions on a linear scale. We note that 1/T_1 ∝ 1/SDW(q)2SDW(q,ω_n), where S(q,ω_n) is the dynamical structure factor. 1/T_1 is a very convenient probe to study the critical dynamics of S(q,ω_n) in the immediate vicinity of magnetic phase transitions because (i) one can probe the dynamics at extremely low energy (ℏω_n ~ μ eV), and (ii) the wave-vector integral is automatically done. Below T_c, we can fit the critical dynamics with a power-law 1/T_1 ∝ (T/T_{SDW} − 1)^−δ. We determined T_{SDW} and the critical exponent δ based on the best fit. The resultant values of T_{SDW} = 99.0 K (Co 2%), 68.9 K (Co 4%), and 42.3 K (Co 5%) are summarized in Fig. 1. The best fit also resulted in the critical exponent δ = 0.329 for Co 2%, and 0.317 for Co 4%. The aforementioned distribution of 1/T_1 below ~70 K for Co 5% makes it difficult to estimate δ with high accuracy, but the observed temperature dependence is consistent with δ ~ 0.33. The inset of Fig. 8 shows a log-log plot of 1/T_1 as a function of the reduced temperature (T/T_{SDW} − 1). The common slope in the vicinity of the SDW transition indicates that the SDW transition of all three compositions belongs to the same universality class, and the critical exponent is given by δ ~ 0.33. This value is close to δ = 0.33 ± 0.01 observed for a Mott-insulator CuO [17] in the vicinity of the Néel transition at T_N = 229 K, and consistent with the prediction for insulating three-dimensional Heisenberg antiferromagnets δ ~ 0.35 [18–21].

C. Ordered moments

In Figs. 2(b) and 2(c), we show the effects of SDW ordering on the field-swept NMR line shapes of Ba(Fe_{0.95}Co_{0.05})_2As_2 with B_{ext} || c. We confirmed the symmetrical nature of the line shape at 4.2 K, as expected, hence only the lower field half of the line shapes was measured in the intermediate temperature range between 4.2 K and T_{SDW}. Below T_{SDW}, the entire 75As NMR line shape begins to split. As noted first by Kitagawa et al. in the case of undoped BaFe_2As_2 [8], this is because the

FIG. 7. (Color online) A semi-log plot of 1/T_1 T observed for Ba(Fe_{1−x}Co_x)2As_2 with B_{ext} || ab. For clarity, we show data points only above T_{SDW} for x ≤ 0.05, and above T_c for x = 0.08 and 0.12. Dashed curves represent a phenomenological Curie-Weiss fit, incorporating a background term due to a pseudogap (see main text). Slanted solid arrows mark T_c for x = 0% ~ 5%, while vertical dashed arrows show T_{SDW} for x = 8% and 12%.

FIG. 8. (Color online) Power-law fits of 1/T_1 in the critical region of underdoped Ba(Fe_{1−x}Co_x)2As_2. Dotted curve and solid arrows mark T_c and T_{SDW}, respectively. Inset: log-log plot of 1/T_1 as a function of the reduced temperature (T/T_{SDW} − 1). Solid lines represent a power-law behavior in the critical region with δ = 0.33.
static hyperfine magnetic field $B_{hf}$ at $^{75}$As sites arising from the ordered Fe moments within the Fe layers points toward the $+c$ or $-c$ axis. For this reason, the overall NMR line shape shifts only slightly without exhibiting a splitting under the configuration of $B_{ext} \parallel ab$, as shown in Fig. 2(d).

While the observed NMR line shapes below $T_{SDW}$ bear similarities with the case of undoped BaFe$_2$As$_2$, there is one major difference [22]: our NMR line shapes in Figs. 2(b) and 2(c) exhibit a continuum in the middle. The integrated intensity between $B_{ext} = 5.474$ to 6.475 T accounts for $\sim$8.5% of the overall intensity. This implies that $\sim$8.5% of $^{75}$As nuclear spins experience $|B_{hf}| \leq 0.5$ T, while the maximum value of the hyperfine field reaches $B_{hf}^{\text{max}} = 1.27$ T at 4.2 K. Our attempt to fit the observed line shape with one-dimensional incommensurate modulation $B_{hf} = B_{hf}^{\text{max}} \sin(\vec{q} \cdot \vec{x})$, where $\vec{q}$ represents the incommensurate SDW ordering vector, is unsatisfactory, as shown in Fig. 2(c). Notice that the calculated results grossly overestimate the spectral weight in the middle part of the line shape. In view of the fact that the integrated intensity of the $^{75}$As(1) sites with a Co atom in one of their four nearest-neighbor Fe sites also accounts for approximately $\sim$7.5% of the intensity [see asterisk in Fig. 2(a)] [26], the continuum in the middle part of the NMR line shape may arise primarily from $^{75}$As(1) sites. That is, Co dopants may be suppressing the Fe magnetic moments locally. It has been shown by neutron scattering that the SDW is commensurate with the lattice up to $x = 0.056$ [42,43]. Based on our NMR data, we can not prove or disprove the incommensurability at $x = 0.02$. We note that similar $^{75}$As line shapes have been observed in the lightly doped regime of Ba(Fe$_{1-x}$Ni$_x$)$_2$As$_2$ ($x = 0.0072$ and 0.016) [24].

We summarize the temperature dependence of $B_{hf}^{\text{max}}$ in Fig. 9. $B_{hf}^{\text{max}}$ remains approximately constant up to $\sim$30 K, then decreases continuously toward $T_{SDW} = 99.0$ K. This behavior is markedly different from the first-order commensurate SDW transition in BaFe$_2$As$_2$ [8]; $B_{hf}$ decreases discontinuously at $T_{SDW} = 135$ K in the latter. By fitting the temperature dependence of $B_{hf}^{\text{max}}$ between $70$ K ($\sim T_{SDW}$) and $T_{SDW} = 99.0$ K to a power law, $B_{hf}^{\text{max}} \sim (T_{SDW} - T)^{\beta}$ with a fixed $T_{SDW} = 99.0$ K, we obtain the critical exponent $\beta \sim 0.3$. Very broad line shapes make accurate determination of $B_{hf}^{\text{max}}$ difficult near $T_{SDW}$, hence we were unable to eliminate the large uncertainties of $\beta$. Nonetheless, it is worth pointing out that $\beta \sim 0.3$ is consistent with the expectation from the Heisenberg model $\beta = 0.37$, but different from the mean-field value $\beta = 0.5$.

Turning our attention to the magnitude of the ordered moment $\mu_{\text{eff}}$ at 4.2 K as a function of $x$, we compare NMR results with those obtained from neutron scattering in Fig. 10. Since $B_{hf}$ has a distribution under the presence of Co dopants, we plot both the maximum value and the center of gravity of the hyperfine field $B_{hf}^{\text{max}}$ and $B_{hf}^{C,G}$, respectively, in Fig. 10(b). We recall that $\mu_{\text{eff}} = 0.87\mu_B$ at 4.2 K for the parent compound BaFe$_2$As$_2$ [6], and Co doping suppresses $\mu_{\text{eff}}$ [41,42], as summarized in Fig. 10(a). On the other hand, $B_{hf}^{\text{max}} - B_{hf}^{C,G} = 1.5$ T observed earlier for BaFe$_2$As$_2$ [8] is gradually suppressed by Co doping. Our results of $B_{hf}^{\text{max}}$ smoothly extrapolate to the critical concentration as determined from the analysis of the $1/T_{1}T$ in Fig. 1, $x_c \sim 6.5$%.

**D. Low-energy spin excitations below $T_{SDW}$**

In Fig. 6, we show the temperature dependence of $1/T_{1}T$ below $T_{SDW}$. Our results show a typical $\lambda$-like temperature dependence in the vicinity of the SDW transition. In insulating antiferromagnets, the low-temperature behavior of $1/T_{1}T$ is usually dominated by multimagnon Raman processes, and $1/T_{1}T$ decreases very quickly [28]. In the present case,
however, as we approach the base temperature of 4.2 K, $1/T_1 T$ levels off to a constant value of $1/T_1 T \sim 0.08 \text{s}^{-1} \text{K}^{-1}$. Analogous behavior was previously reported also for the undoped parent phase BaFe$_2$As$_2$, and was attributed to the Korringa process arising from low-energy electron-hole pair excitations at the reconstructed Fermi surface [8]. We summarize the values of $1/T_1 T$ observed at 4.2 K as a function of the doping content $x$ in the inset of Fig. 6, including our preliminary results for $x = 0.04$ [22]. Interestingly, three data points fit nicely with a parabolic function of $x$. If the sizable magnitude of $1/T_1 T$ at 4.2 K indeed arises from the Korringa process, we expect the observed parabolic increase of $1/T_1 T$ implies that $D(E_F)$ increases roughly linearly with $x$. We note that if we apply a simple rigid band picture to the reconstructed Fermi surfaces, simple dimensional analysis of $E_F$ and $D(E_F)$ in three dimensions would lead to $D(E_F) \propto x^{1/3}$ instead, where $x$ is the number of conduction electrons.

IV. SUMMARY AND CONCLUSIONS

We have presented an in-depth $^{75}$As NMR study of the critical behavior of the SDW transition in the lightly Co-doped regime of Ba(Fex$_{0.1}$Co$_{0.9}$)$_2$As$_2$, with the primary focus on $x = 0.02$. We identified the NMR signatures of the tetragonal to orthorhombic structural phase transition preceding the SDW transition. Our Knight shift, NMR linewidth, and $1/T_1$ data suggest that the strong short-range SDW order with three-dimensional nature sets in once the FeAs planes lower the symmetry from tetragonal to orthorhombic. In the orthorhombic phase below $T_c$, simplistic fits of the antiferromagnetic contribution to $1/T_1 T$ based on a Curie-Weiss law using two free parameters (Fig. 7) or 2D SCR (self-consistent renormalization) theory with four free parameters [44] fail to capture the critical behavior. Precisely at $T_c$, critical slowing down of spin fluctuations sets in, and the critical exponent for the divergence of the dynamical structure factor $S(q, \omega_n)$ is $\delta \sim 0.33$, as generally expected for insulating 3D Heisenberg antiferromagnets. Our fitting range is rather limited and it is difficult to draw a definitive conclusion, but this value is inconsistent with $\delta = 0.5$ expected for the 3D SCR theory for itinerant antiferromagnets [45].

ACKNOWLEDGMENTS

The work at Zhejiang was supported by National Basic Research Program of China (Grants No. 2014CB921203 and No. 2011CB00103), NSF of China (Grant No. 11274268). The work at McMaster was supported by NSERC and CIFAR. The work at Oak Ridge National Laboratory was supported by the Department of Energy, Basic Energy Sciences, Materials Sciences and Engineering Division. The work at Beijing and Nanjing was supported by NSFC, the Ministry of Science and Technology of China, and the Chinese Academy of Sciences.

[1] X. H. Chen, T. Wu, G. Wu, R. H. Liu, H. Chen, and D. F. Fang, Nature (London) 453, 761 (2008).
[2] Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, J. Am. Chem. Soc. 130, 3296 (2008).
[3] Z. A. Ren, W. Lu, J. Yang, W. Yi, X. L. Shen, Z. C. Li, G. C. Che, X. L. Dong, L. L. Sun, F. Zhou, and Z. X. Zhao, Chin. Phys. Lett. 25, 2215 (2008).
[4] M. Rotter, M. Tegel, and D. Johrendt, Phys. Rev. Lett. 101, 107006 (2008).
[5] A. S. Sefat, R. Jin, M. A. McGuire, B. C. Sales, D. J. Singh, and D. Mandrus, Phys. Rev. Lett. 101, 117004 (2008).
[6] 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).
[7] H. Fukazawa, K. Hirayama, K. Kondo, T. Yamazaki, Y. Kohori, N. Takeshita, K. Miyazawa, H. Kito, H. Eisaki, and A. Iyo, J. Phys. Soc. Jpn. 77, 093706 (2008).
[8] K. Kitagawa, N. Katayama, K. Ohgushi, M. Yoshida, and M. Takigawa, J. Phys. Soc. Jpn. 77, 114709 (2008).
[9] N. Mi, M. E. Tillman, J. Q. Yan, A. Kracher, S. T. Hannahs, S. L. Bud’ko, and P. C. Canfield, Phys. Rev. B 78, 214515 (2008).
[10] F. L. Ning, K. Ahilan, T. Imai, A. S. Sefat, R. Jin, M. A. McGuire, B. C. Sales, and D. Mandrus, J. Phys. Soc. Jpn. 78, 013711 (2009).
[11] J. H. Chu, J. G. Analytis, C. Kucharczyk, and I. R. Fisher, Phys. Rev. B 79, 014506 (2009).
[12] S. Nandi, M. G. Kim, A. Kreyssig, R. M. Fernandes, D. K. Pratt, A. Thaler, N. Ni, S. L. Bud’ko, P. C. Canfield, J. Schmalian, R. J. McQueeney, and A. I. Goldman, Phys. Rev. Lett. 104, 057006 (2010).
[13] X. F. Wang, T. Wu, J. Wu, R. H. Liu, X. H. Chen, and Y. L. Xie, New J. Phys. 11, 045003 (2009).
[14] L. Fang, H. Luo, P. Cheng, Z. Wang, Y. Jia, G. Mu, B. Shen, I. M. Mazin, L. Shan, C. Ren, and H.-H. Wen, Phys. Rev. B 80, 140508(R) (2009).
[15] D. C. Johnston, Adv. Phys. 59, 803 (2010).
[16] K. Kontani and Y. Masuda, J. Magn. Magn. Mater. 31-34, 287 (1993).
[17] Y. Itoh, T. Imai, T. Shimizu, T. Tsuda, H. Yasuoka, and Y. Ueda, J. Phys. Soc. Jpn. 59, 1143 (1990).
[18] B. I. Halperin and P. C. Hohenberg, Phys. Rev. Lett. 19, 700 (1967).
[19] C. Hohenemser, N. Rosov, and A. Kleinhammes, Hyperfine Interact. 49, 267 (1989).
[20] K. Kawasaki, Prog. Theor. Phys. 39, 285 (1968).
[21] S. W. Lovesey, E. Balcar, and A. Cuccoli, J. Phys.: Condens. Matter 7, 2615 (1995).
[22] F. L. Ning, K. Ahilan, T. Imai, A. S. Sefat, R. Jin, M. A. McGuire, B. C. Sales, and D. Mandrus, Phys. Rev. B 79, 140506(R) (2009).
[23] A. P. Dioguardi, J. Crocker, A. C. Shockley, C. H. Lin, K. R. Shiner, D. M. Nisson, M. M. Lawson, N. apRoberts-Warren, P. C. Canfield, S. L. Bud’ko, S. Ran, and N. J. Curro, Phys. Rev. Lett. 111, 207201 (2013).
[24] A. P. Dioguardi, N. apRoberts-Warren, A. C. Shockley, S. L. Bud’ko, N. Ni, P. C. Canfield, and N. J. Curro, Phys. Rev. B 82, 140411(R) (2010).
[25] F. Hammerath, U. Gräfe, T. Kühne, H. Kühne, P. L. Kuhns, A. P. Reyes, G. Lang, S. Wurmehl, B. Büchner, P. Carretta, and H.-J. Grafe, Phys. Rev. B 88, 104503 (2013).

[26] F. L. Ning, K. Ahilan, T. Imai, A. S. Sefat, M. A. McGuire, B. C. Sales, D. Mandrus, P. Cheng, B. Shen, and H. H. Wen, Phys. Rev. Lett. 104, 037001 (2010).

[27] F. L. Ning, K. Ahilan, T. Imai, A. S. Sefat, R. Jin, M. A. McGuire, B. C. Sales, and D. Mandrus, J. Phys. Soc. Jpn. 77, 103705 (2008).

[28] V. Jaccarino, in Proceedings of the International School of Physics, Enrico Fermi XXXVII (Academic, New York, 1967).

[29] S. Oh, A. M. Mounce, S. Mukhopadhyay, W. P. Halperin, A. B. Vorontsov, S. L. Bud’ko, P. C. Canfield, Y. Furukawa, A. P. Reyes, and P. L. Kuhns, Phys. Rev. B 83, 214501 (2011).

[30] T. Imai, C. P. Slichter, K. Yoshimura, and K. Kosuge, Phys. Rev. Lett. 70, 1002 (1993).

[31] R. M. Fernandes, A. E. Böehmer, C. Meingast, and J. Schmalian, Phys. Rev. Lett. 111, 137001 (2013).

[32] R. M. Fernandes, L. H. VanBebber, S. Bhattacharya, P. Chandra, V. Keppens, D. Mandrus, M. A. McGuire, B. C. Sales, A. S. Sefat, and J. Schmalian, Phys. Rev. Lett. 105, 157003 (2010).

[33] M. Yoshizawa, D. Kimura, T. Chiba, A. Ismayil, Y. Nakamishi, K. Kihou, C. H. Lee, A. Iyo, H. Eisaki, M. Nakajima, and S. Uchida, J. Phys. Soc. Jpn. 81, 024604 (2012).

[34] A. Smerald and N. Shannon, Phys. Rev. B 84, 184437 (2011).

[35] 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).

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

[37] D. A. Torchetti, M. Fu, D. C. Christensen, K. J. Nelson, T. Imai, H. C. Lei, and C. P. Petrovic, Phys. Rev. B 83, 104508 (2011).

[38] Y. Nakai, K. Ishida, Y. Kamihara, M. Hirano, and H. Hosono, J. Phys. Soc. Jpn. 77, 073701 (2008).

[39] R. Zhou, Z. Li, J. Yang, C. T. Lin, and G. Q. Zheng, Nat. Commun. 4, 2265 (2013).

[40] M. Fu, D. A. Torchetti, T. Imai, F. L Ning, J.-Q. Yan, and A. S. Sefat, Phys. Rev. Lett. 109, 247001 (2012).

[41] C. Lester, Jiun-Haw 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).

[42] 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. McQueeney, Phys. Rev. Lett. 103, 087001 (2009).

[43] D. K. Pratt, M. G. Kim, A. Kreyssig, Y. B. Lee, G. S. Tucker, A. Thaler, M. Tian, J. L. Zarestky, S. L. Bud’ko, P. C. Canfield, B. N. Harmon, A. I. Goldman, and R. J. McQueeney, Phys. Rev. Lett. 106, 257001 (2011).

[44] Y. Nakai, T. Iye, S. Kitagawa, K. Ishida, S. Kasahara, T. Shibauchi, Y. Matsuda, H. Ikeda, and T. Terashima, Phys. Rev. B 87, 174507 (2013).

[45] T. Moriya, Spin Fluctuations in Itinerant Electron Magnetism (Springer, New York, 1985).