Normal state spin dynamics in the iron-pnictide superconductors 
$\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)2$ and $\text{Ba(Fe}_{1-x}\text{Co}_x)2\text{As}_2$ probed with NMR measurements

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(Dated: May 14, 2013)

The NMR results in iron pnictides $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)2$ and $\text{Ba(Fe}_{1-x}\text{Co}_x)2\text{As}_2$ are analyzed based on the self-consistent renormalization (SCR) spin fluctuation theory. The temperature dependence of the NMR relaxation rate $T_1^{-1}$ as well as the electrical resistivity is well reproduced by a SCR model where two-dimensional antiferromagnetic (AF) spin fluctuations are dominant. The successful description of the crossover feature from non-Fermi liquid to Fermi liquid behavior strongly suggests that low-lying spin fluctuations in $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)2$ and $\text{Ba(Fe}_{1-x}\text{Co}_x)2\text{As}_2$ possess an itinerant AF nature, and that chemical substitution in the two compounds tunes the distance of these systems to an AF quantum critical point. The close relationship between spin fluctuations and superconductivity is discussed compared with the other unconventional superconductors, cuprate and heavy fermion superconductors. In addition, it is suggested that magnetism and lattice instability in these pnictides are strongly linked via orbital degrees of freedom.

PACS numbers: 76.60.-k, 74.25.nj, 74.40.Kb, 74.70.Xa

I. INTRODUCTION

Since the discovery of high-temperature superconductivity in iron-pnictide superconductors, much effort has been paid to the understanding of the normal and superconducting (SC) state properties, and considerable interest has been focused on the origin of the pairing interaction. The proximity of a SC to an antiferromagnetic (AF) quantum critical point (QCP) is suggested to be close to an AF quantum critical point (QCP) on the basis of the self-consistent renormalization (SCR) theory of spin fluctuations. The SCR theory, developed by Moriya and coworkers, has been applied for weak ferromagnetism and antiferromagnetism of $d$-electron itinerant magnets, and succeeded in characterizing properties of spin fluctuations. As recent studies have shown the importance of both the itinerant and localized nature of the magnetism of iron pnictides, it is important to show to what extent experimental results are understood within an itinerant and local-moment picture. Recently, x-ray emission spectroscopy, which is sensitive to very rapid time scales, allowed for the detection of large local moments in the paramagnetic states in iron pnictides. In contrast, NMR is a very useful probe to detect much slower fluctuations or low-energy spin excitation, enabling us to extract the itinerant aspects of iron pnictides.

We derive spin-fluctuation parameters in the two compounds by taking into account other experimental results such as the static magnetic susceptibility and specific heat. We calculate the temperature dependencies of the NMR relaxation rate and the electrical resistivity following the SCR theory, and show that our calculations are qualitatively consistent with the experimental data. Our analysis indicates that the $T_c$ maximum concentration corresponds to an AF QCP and suggests the possibility of magnetically mediated high-$T_c$ superconductivity in the “122” iron-pnictide superconductors as in other unconventional superconductors of strongly-correlated-electron systems.

In this paper, we analyze in more detail experimental results, particularly the NMR relaxation rate of $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)2$ and $\text{Ba(Fe}_{1-x}\text{Co}_x)2\text{As}_2$. They both possess the “122” structure (ThCr$_2$Si$_2$ structure) and the recent thermal expansion experiment showed their thermodynamic similarity. These compounds are suggested to be close to an AF quantum critical point (QCP) on the basis of the self-consistent renormalization (SCR)
II. SURVEY OF NMR EXPERIMENTS

Most of our NMR experimental results in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ were published previously. In order to reanalyze our published NMR data in terms of the SCR theory, we summarize them in Figs. 1 and 2.

Figure 1 demonstrates the temperature and P-concentration dependence of the Knight shift in BaFe$_2$(As$_{1-x}$P$_x$)$_2$, which is a measure of the static spin susceptibility $\chi(q=0)$. The Knight shift is basically $T$-independent, but P substitution reduces the magnitude of the Knight shift. These results are attributable to the decrease in the density of states (DOS) at the Fermi level with P substitution.

Figure 2 displays the temperature dependence of $(T_1 T)^{-1}$ for BaFe$_2$(As$_{1-x}$P$_x$)$_2$ with various P-concentration, where P substitution suppresses antiferromagnetism and induces superconductivity. We observe non-Fermi-liquid (NFL) temperature dependence of the Curie-Weiss (CW) form $(T_1 T)^{-1} = a + b/(T + \theta)$ in the paramagnetic temperature range. For $x \leq 0.20$, $(T_1 T)^{-1}$ increases on cooling and has a peak at $T_N$ due to the opening of spin density wave gap, but for $x \geq 0.33$, $(T_1 T)^{-1}$ exhibits a peak due to a SC gap opening. The CW-type temperature dependence indicates the presence of two-dimensional (2D) AF spin fluctuations according to the SCR theory. The crossover from Fermi-liquid to CW behavior in $(T_1 T)^{-1}$ correlates perfectly with the change in the resistivity results. As the system evolves from a Fermi liquid ($x = 0.71$) towards the maximum $T_c$ ($x = 0.33$) near the AF phase, the temperature dependence of the resistivity changes from $T^2$ to $T$ linear, one hallmark of NFL behavior.

We show in the next section that the CW behavior of $(T_1 T)^{-1}$ is consistent with the observed temperature dependences of the electrical resistivity $\rho$ and with the predictions of a SCR model with spin-fluctuation parameters relevant to BaFe$_2$(As$_{1-x}$P$_x$)$_2$.

III. ANALYSIS BASED ON THEORY OF SPIN FLUCTUATIONS

In this section, we demonstrate that experimental data of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ and Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ are quantitatively explainable in terms of the SCR theory for two-dimensional itinerant antiferromagnets. All the NMR data of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ are cited from Ref. 6.

A. Outline of the self-consistent renormalization (SCR) theory

The SCR theory gives quantitative relations between dynamical susceptibility and physical properties. In nearly and weakly AF metals, dynamical susceptibility above $T_N$ for a wave vector near the AF ordering vector $Q$ may generally be written as follows:

$$\chi(Q + q, \omega) = \frac{\chi(Q + q)}{1 - i\omega/\Gamma_{Q+q}}$$

with

$$[\chi(Q + q)]^{-1} = [\chi(Q)]^{-1} + Aq^2,$$
$$\Gamma_{Q+q} = \Gamma(\kappa^2 + q'^2),$$
$$\kappa^2 = 1/\chi(Q),$$

where $\kappa^{-1}(= \xi_T)$ is the temperature-dependent magnetic correlation length, and $A$ and $\Gamma$ are temperature independent constants, which are the fundamental parameters of the theory. Using above relations, the dynamical spin
susceptibility \( \chi''(Q, \omega) \) is written as

\[
\chi''(Q + q, \omega) = \chi(Q + q) \frac{\omega \Gamma q^2}{\omega^2 + \Gamma q^2 Q + q^2} \tag{1}
\]

\[
= \chi(Q) \kappa^2 \frac{\omega \Gamma (\kappa^2 + q^2)}{\kappa^2 + q^2 \omega^2 + [\Gamma (\kappa^2 + q^2)]^2} \tag{2}
\]

\[
= \frac{\Gamma}{\kappa^2 + q^2} \frac{\omega}{\omega^2 + [\Gamma (\kappa^2 + q^2)]^2} \tag{3}
\]

\[
= \frac{\Gamma}{\kappa^2 + q^2} \frac{\omega}{\omega^2 + [\Gamma (\kappa^2 + q^2)]^2} \tag{4}
\]

\[
= \frac{\pi T_0}{T_A} \omega + \left( 2 \pi T_0 y + (q/q_B)^2 \right) \frac{\omega}{\omega^2 + [\Gamma (\kappa^2 + q^2)]^2} \tag{5}
\]

where \( q_B \) is the cut-off wave vector and has a relation of \( s_0 q_B^2 / 4\pi = 1 \) with \( s_0 \) being the area per magnetic atom in the 2D plane. In this formula, important spin-fluctuation parameters are the following two characteristic temperatures,

\[
T_0 = \frac{\Gamma q_B^2}{2\pi}
\]

\[
T_A = \frac{A q_B^2}{2\pi},
\]

which characterize the width of the spin excitations spectrum in frequency \( \omega \) and momentum \( q \) space. The dimensionless inverse susceptibility \( y(T) \) at \( Q = Q_{AF} \) of AF wave vectors is defined as

\[
y(T) = \frac{\kappa^2}{q_B^2} = \frac{1}{A \chi(Q) q_B^2} = \frac{1}{2T_A \chi(Q)}. \tag{6}
\]

Here, \( y_0 \) is the zero temperature limit of \( y \), and characterizes the proximity to the magnetic instability. \( y_0 = 0 \) indicates an AF QCP, where \( \chi(Q) \) diverges down to zero temperature.

The staggered susceptibility or \( y \) is determined self-consistently from the relation of the mean square local amplitude of the zero point and thermal spin fluctuations, and is calculated from

\[
y = y_0 + \frac{y_1 t}{2} \left( \phi(y/t) - \phi(y/t + 1/t) \right), \tag{7}
\]

where \( t = T/T_0 \), \( y_1 \) is the parameter which governs the mode-mode coupling of AF spin fluctuations, and \( \phi(x) \) is given as,

\[
\phi(x) = \left( x - \frac{1}{2} \right) \log x + x \log \Gamma(x) - \log \sqrt{2\pi}. \tag{8}
\]

B. Calculations of the nuclear spin-lattice relaxation rate \( 1/T_1 \)

Nuclear spin-lattice relaxation rate \( 1/T_1 \) is generally expressed by

\[
\frac{1}{T_1} = \gamma_N^2 T \lim_{\omega \to 0} \sum_q \frac{|A_q|^2 \chi''(q, \omega_0)}{\omega_0} \tag{9}
\]

where \( \gamma_N \) is the gyromagnetic ratio of an observed nucleus, \( N_A \) is the number of magnetic atoms per unit volume, \( A_q \) is the coupling constant for the hyperfine interaction between the nuclear spin and the \( q \)-component of the spin density, \( \omega_0 \) is the NMR frequency (order of milliKelvin). Inserting Eq. (5) into Eq. (3) and neglecting the \( q \) dependence of \( A_q \), \( T_1^{-1} \) is described as follows

\[
T_1^{-1} = \frac{2\gamma_N^2 A_B^2 T}{N_A} \sum_q \frac{\pi T_0}{\sqrt{2\pi}} \left[ \left( \frac{\pi T_0 y + (q/q_B)^2}{\sqrt{2\pi}} \right) \right] \tag{10}
\]

\[
= \frac{2A_B^2 T}{\pi T_A} \bar{T}_1^{-1} \tag{11}
\]

\[
\bar{T}_1^{-1} = \frac{2}{T_0} \int_0^1 dx \frac{x}{y + x^2} = t \left( \frac{1}{y} - \frac{1}{y + 1} \right), \tag{12}
\]

where \( A_B \) is the hyperfine coupling constant. Thus, \( 1/T_1 \) directly measures the temperature dependence of \( \chi(Q) = [2T_{AF}(T)]^{-1} \).

C. Calculations of the electrical resistivity

In the framework of the SCR theory, predominant contribution to the resistivity arises from the spin fluctuations with AF wave vectors around \( Q_{AF} \). The electrical resistivity in an electron system scattered by those spin fluctuations is calculated based on Boltzmann equation and is given by

\[
R(T) = r R(T) \tag{13}
\]

\[
R(T) = t \left[ \phi \left( \frac{y}{t} \right) - \phi \left( \frac{(y + 1)}{t} \right) \right] \tag{14}
\]

\[
+ y \left[ \log \left( \frac{y}{t} \right) - \psi \left( \frac{y}{t} \right) \right] - \left( y + 1 \right) \left[ \log \left( \frac{(y + 1)}{t} \right) - \psi \left( \frac{(y + 1)}{t} \right) \right]
\]

where \( r \) is an adjustable fitting constant which represents the coupling between the spin fluctuations and conduction electrons, and \( \psi(x) \) is the digamma function.

Linear temperature dependence of the resistivity is generic to a QCP of 2D AF metals. Away from the QCP the electrical resistivity shows a crossover from the anomalous \( T \)-linear dependence to the Fermi-liquid like \( T^2 \) behavior.

D. Analysis on spin fluctuations in \( Ba(Fe_{1-x}Co_x)_2As_2 \)

Inelastic neutron scattering measurements revealed that two-dimensional spin fluctuations possess the stripe correlations \( |Q_{AF}| = (0, \pi) \) or \( (\pi, 0) \) in an unfolded Brillouin zone. The presence of the stripe correlations is also suggested from the anisotropy of NMR \( 1/T_1 \) at the As site. The \( T_1 \) anisotropic ratio \( R = \frac{T_1}{T_1^{H \perp}} \) at the As site \( \sim 1.5 \) above \( T_N \) in BaFe \( _2 \) As \( _2 \).
SrFe$_2$As$_2$ and LaFeAsO is consistently understood from the anisotropic spin fluctuations due to the off-diagonal components ($B_{ac}$) of hyperfine coupling tensor $B$ at the As site and from the stripe correlations of the Fe spins. The importance of the off-diagonal terms was first pointed out by Kitagawa et al. in the internal magnetic fields produced by diagonal terms ($B_{aa}$) are canceled out even if the spin correlations are stripe, since As atoms are located at the symmetrical site with respect to the four nearest neighbor Fe atoms. The off-diagonal terms related with the stripe correlations are discussed later.

Eqs. (11) and (12) only give the contribution of spin fluctuations around the AF wave vector $Q_{AF}$. Although the AF contribution around $Q_{AF}$ is expected to be predominant for NMR relaxation rate in BaFe$_2$(As$_{1-x}$P$_x)_2$ and Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$, there is an additional contribution arising from spin fluctuations around $q = 0$. The observed spin-lattice relaxation rate is thus decomposed into the following two components:

$$
\left(\frac{1}{T_1T}\right)_{\text{obs.}} = \left(\frac{1}{T_1T}\right)_{q \sim 0} + \left(\frac{1}{T_1T}\right)_{q \sim Q_{AF}}.
$$

The experimental NMR results of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ reported by Ning et al. show significant AF fluctuations near the optimal doping of $x \sim 0.06$, and that the AF spin fluctuations are systematically suppressed by Co doping. In addition, they reported that $(T_1T)^{-1}$ decreases on cooling for over-doped samples, as observed in LaFeAsO(O,F) indicating that AF spin stripe correlations are not significant in highly over-doped samples. These results are in good agreement with the inelastic neutron scattering measurements. Since the NMR results of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ indicate that the background term of $(T_1T)^{-1}$, which is ascribable to $(T_1T)^{-1}_{q \sim 0}$, shows non-monotonic behavior, the analysis of contributions from AF fluctuations is less straightforward than BaFe$_2$(As$_{1-x}$P$_x)_2$, which will be shown below. By assuming the temperature dependence of the background term of $(T_1T)^{-1}$ is identical with that of the Knight shift, Ning et al. estimated the AF contribution and found that its temperature dependence follows Curie-Weiss-type $(T_1T)^{-1}_{q \sim Q_{AF}} = C/(T + \theta)$ as observed in BaFe$_2$(As$_{1-x}$P$_x)_2$. They thus employed the following phenomenological two-component model:

$$(T_1T)^{-1}_{\text{obs.}} = (T_1T)^{-1}_{q \sim Q_{AF}} + (T_1T)^{-1}_{q \sim 0}$$

$$
(T_1T)^{-1}_{q \sim 0} = \alpha K_{\text{spin}} = \alpha (a + b \exp(-\Delta/k_BT)).
$$

By using their estimation of $(T_1T)^{-1}_{q \sim 0}$, the doping dependence of $(T_1T)^{-1}_{q \sim Q_{AF}}$, was obtained as shown in Fig. 3. Since $(T_1T)^{-1} = \text{const.}$ behavior is an indication of the verge of a 2D AF QCP (see the next section E), we expect a critical Co concentration of 0.05 $< x < 0.08$ in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$.

For simulating the NMR data, we need to determine $y_0$, $y_1$, $T_0$, and $T_A$. In order to narrow down SCR parameters, we analyzed inelastic neutron scattering data. Inosov et al. reported that the temperature dependence of the damping constant $\Gamma(T)$ of the dynamical spin susceptibility for nearly optimally doped Ba(Fe$_{0.925}$Co$_{0.075}$)$_2$As$_2$ ($T_c = 25$ K) shows a linear temperature dependence $\Gamma(T) = 0.14(T + 30)[\text{meV}]$. The $\Gamma(T)$ can be calculated in the SCR theory as follows:

$$
\Gamma(T) = 2\pi T_0y(T).
$$

FIG. 3: (Color online) The $T$-dependence of the NMR relaxation rate arising from the $q \sim Q_{AF}$ mode of spin fluctuations $(T_1)_{\text{AF}}$ of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ for $H \parallel ab$, cited from Ref. 6. The solid lines represent simulations with the SCR parameters listed in Table I.
specific heat as follows. In the framework of the SCR theory, we can relate the magnetic susceptibility to $T_A$ using the following relation:\cite{33,37}

$$T_A = 0.75/\chi \text{(in emu/mol).} \quad (19)$$

The estimated $T_A$ of $x = 0.08$ from the NMR and neutron scattering data corresponds to the susceptibility at $T \approx 200$ K.\cite{35} For other $x$ values, we thus use the $T_A$ values estimated from the susceptibility at 200 K for our calculation.\cite{35} The doping dependence of $T_A$ is estimated from the reported specific heat experiment\cite{36} by using the following relation:\cite{33,37}

$$\gamma = \frac{6200}{T_0} (2x_c - \pi y_0)^{-1/2} \quad [\text{mJ/mol K}^2]. \quad (20)$$

where $x_c$ is the cut-off wave vector of which magnitude is the order of unity. Note that we used the first term for $y_0 < 0$.\cite{33}

In this way, we simulate the Co concentration dependence of the NMR relaxation rate as shown in Fig. 6, with the SCR parameters as listed in Table I indicating very good agreement with the experimental data. Using the same parameter, we also calculated the temperature dependence of the electrical resistivity and found good agreement with the experimental result as shown in Fig. 5.

$\begin{array}{|c|c|c|}
\hline
x & y_0 & T_0 (K) & T_A (K) \\
\hline
0.00 & -0.4 & 8.0 & 460 & 800 \\
0.04 & -0.07 & 3.0 & 510 & 950 \\
0.05 & -0.03 & 2.7 & 530 & 960 \\
0.08 & 0.025 & 2.5 & 450 & 1050 \\
0.1 & 0.05 & 4.0 & 420 & 1200 \\
\hline
\end{array}$

TABLE I: The SCR parameters of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$.

FIG. 5: The $T$-dependence of the electrical resistivity $\rho_{ab}$ of Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$, cited from Ref. 27. The solid line represents a calculation using $y_0 = 0.025$, $y_1 = 2.5$, and $T_0 = 450$ K, and residual resistivity $\rho_{ab}^0 = 0.1$ mΩcm.

FIG. 6: (Color online) The $T^{-1}$ of $^{31}$P and $^{75}$As in BaFe$_2$(As$_{1-x}$P)$_2$ with $x = 0.33$ is plotted. We estimated the hyperfine coupling constant of $^{31}$P nucleus from the slope as $A_{hf} = 0.674$ T/$\mu_B$ by using $^{75}$As $A_{hf} = 1.72$ T/$\mu_B$.\cite{36}

E. Analysis on spin fluctuations in BaFe$_2$(As$_{1-x}$P)$_2$

Following the similar procedure as in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$, we here analyze the experimental NMR results of BaFe$_2$(As$_{1-x}$P)$_2$. Because the uniform susceptibility of BaFe$_2$(As$_{1-x}$P)$_2$ is almost temperature independent, the NMR relaxation rate arising from the small-$q$ fluctuations $(T)_q^{-1}$ is also expected to be temperature independent. In order to estimate $(T)_q^{-1}$, we considered the CW-type equation: $(T)_q^{-1} = a + b/(T + \theta)$. We relate the first constant term $a$ with the $(T)_q^{-1}$ and estimated $(T)_q^{-1}$ as shown in Fig. 6. The nearly constant $(T)_q^{-1}$ of $x = 0.33$ suggests that $y_0$ is very close to zero at $x = 0.33$.\cite{33}

For simulations of $T^{-1}$, one needs the hyperfine coupling constant $A_{hf}$ of $^{31}$P nucleus. In order to estimate it, it is reasonably assumed that the $T^{-1}$ of $^{31}$P is determined by the off-diagonal terms of the hyperfine coupling tensor, as is the case for the $T^{-1}$ of $^{75}$As in BaFe$_2$As$_2$.\cite{27} Figure 6 displays the $T^{-1}$ of $^{75}$As for $x = 0.33$ plotted against that of $^{31}$P with temperature as an implicit parameter. Since the $T^{-1}$ of $^{31}$P is proportional to that of $^{75}$As as shown in Fig. 6, we can estimate $^{31}A_{hf} = 6.37$ kOe/$\mu_B$ for $^{31}$P nucleus by using $^{75}A_{hf} = 17.2$ kOe/$\mu_B$ for $^{75}$As nucleus.\cite{27} We also assume in our calculations that the hyperfine coupling constant is independent of P concentration.

Although complete P concentration dependence of magnetic susceptibility and specific heat is not reported in BaFe$_2$(As$_{1-x}$P)$_2$, we estimate $T_0$ and $T_A$ as follows. The characteristic spin fluctuation energy $T_0$ of $x = 0.33$ with $y_0 = 0$ can be estimated from reported specific heat experiments\cite{14} by using Eq. (20). Assuming that the P concentration dependence of $\gamma$ is identical with that of $K_{sp}$ which is the measure of the DOS at the Fermi level.
energy we can estimate γ for other P concentrations and obtain the P concentration dependence of T0 using Eq. (20). We here neglect the second term in Eq. (20) for simplicity. In order to estimate T_A from Eq. (14), we assume the magnetic susceptibility χ is proportional to K_{spin}. By using χ = 9.4 × 10^{-4} emu/mol at 200 K and γ = 27 mJ/molK^2 in BaFe2As2 35,36 we thus estimate the P concentration dependence of T_A.

By using the SCR parameters listed in Table III which are obtained from our NMR simulation shown in Fig. 7, we calculated the temperature exponent of electrical resistivity. For 2D AF fluctuations, a T-linear resistivity is expected near the QCP. 35 Away from the QCP, the temperature dependence of the resistivity crossovers to a Fermi-liquid-like T^2 as T decreases. The experimental data is actually consistent with the simulated temperature dependence as shown in Fig. 8.

According to the SCR theory, we can also estimate the in-plane spin correlation length ξ(T) and the damping constant Γ(T) from (√4πy)^{-1} and from 2πT0y, respectively. 30 We calculated ξ/a and Γ(T) at T_c for different P concentration as shown in Table III which may be confirmed by future neutron scattering experiments.

| x   | y | y0 | T0 (K) | T0c (K) | ξ(T0)/a (meV) | Γ(T0) | y0 | y1 | T0 | T0c | ξ(T0)/a | Γ(T0) |
|-----|---|----|--------|---------|---------------|--------|----|----|----|-----|---------|--------|
| 0   | 0.0 | 8.0 | 460    | 460     | 3.0           | 3.7    | 0.0 | 8.0 | 460 | 460 | 3.0    | 3.7    |
| 0.2 | -0.15 | 15 | 760    | 1320    |               |        | 0.2 | -0.15 | 15 | 760 | 1320   |        |
| 0.25 | -0.05 | 10 | 770    | 1340    |               |        | 0.25 | -0.05 | 10 | 770 | 1340   |        |
| 0.33 | 0 | 8.0 | 780    | 1350    | 1.9           | 9.2    | 0.33 | 0 | 8.0 | 780 | 1350   | 1.9    |
| 0.41 | 0.06 | 5.0 | 800    | 1390    | 1.1           | 27     | 0.41 | 0.06 | 5.0 | 800 | 1390   | 1.1    |
| 0.56 | 0.2 | 6.0 | 850    | 1480    | 0.6           | 96     | 0.56 | 0.2 | 6.0 | 850 | 1480   | 0.6    |

TABLE II: The SCR parameters of BaFe2(As1−xP_x)2. The in-plane spin correlation length ξ/a and damping constant Γ at T_c are also shown. Note that a is the in-plane lattice constant.

IV. DISCUSSION

A. Phase diagrams

The phase diagrams of BaFe2(As1−xP_x)2 and Ba(Fe1−xCo_x)2As2 are plotted in Fig 9. The y_0 increases with chemical substitution from a negative value in BaFe2As2 to nearly zero around an optimal concentration: x ∼ 0.3 for BaFe2(As1−xP_x)2 and x ∼ 0.06 for Ba(Fe1−xCo_x)2As2. Since y_0 is a measure of the closeness to a QCP, this indicates that their optimal concentration corresponds to an AF QCP and the closeness to the QCP is controllable by P and Co substitution.

B. Spin fluctuation temperature T_0c, T_A versus SC transition temperature T_c

In BaFe2(As1−xP_x)2 and Ba(Fe1−xCo_x)2As2, the SC phase exists next to the AF phase, and T_c is maximum nearly at an AF QCP, i.e. y_0 ∼ 0 as shown in Fig. 9. This strongly suggests that there is an intimate link between superconductivity and antiferromagnetism in iron pnictide superconductors. This is reminiscent of heavy fermion (HF) superconductors, particularly Ce-based superconductors such as CeCu2Si2 and CeMIn5 (M: Co, Rh, and Ir). 37,38 In these Ce-based HF superconductors, superconductivity occurs near an AF QCP, which is induced by competition between the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and the Kondo effect. A number of experiments in these HF superconductors reported NFL behavior (e.g. ρ(T) ∝ T^-α over a wide T range at low temperatures) near the QCP. The NFL behavior is ascribed to AF spin fluctuations with a quantum critical nature, and the AF spin fluctuations

FIG. 7: (Color online) The experimental and calculated NMR relaxation rate arising from q ∼ Q_{AF} mode of spin fluctuations in BaFe2(As1−xP_x)2. The data points represent the experimental data, while the solid lines indicate the calculated data using the SCR parameters listed in Table III.

FIG. 8: (Color online) The experimental and calculated electrical resistivity of BaFe2(As1−xP_x)2. The solid lines represent the calculated data, while the dotted lines indicate the calculated data using the SCR parameters listed in Table III.
SC
AFM
FIG. 9: (Color online) Phase diagrams of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ and Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$, where $T_N$, $T_c$ denotes an AF transition temperature, SC transition temperature, respectively. In the both materials, the concentration where $T_c$ peaks ($x \sim 0.3$ for BaFe$_2$(As$_{1-x}$P$_x$)$_2$, and $x \sim 0.06$ for Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$) exists near the region where $y_0 = 0$. Since $y_0 = 0$ corresponds to an AF QCP, these phase diagrams suggest a close link between superconductivity and AF quantum criticality.

likely induce unconventional superconductivity with a $d$-wave order parameter. Similarly, AF fluctuations are also suggested for a likely candidate of the pairing mechanism for high-$T_c$ cuprate superconductors where significant NFL behavior is observed, although understanding of the pseudogap behavior in the normal state has not been settled.

In order to understand a relationship between AF spin fluctuations and superconductivity, we plot SC $T_c$ against spin-fluctuation parameters, $T_0$ and $T_A$ of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ and Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ as well as those of unconventional superconductors in Fig. 10. Note that only optimal BaFe$_2$(As$_{1-x}$P$_x$)$_2$ and nearly optimal Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ are plotted, since the (nearly) optimal samples are close to an AF QCP. The linear scaling between the spin fluctuation temperature $T_0$ and $T_c$ in Ce-based HF superconductors and the cuprates was interpreted as an indication of spin-fluctuation mediated superconductivity in these unconventional superconductors and that a higher spin fluctuation temperature can give rise to a pairing interaction and thus resulting in higher $T_c$. Interestingly, optimal BaFe$_2$(As$_{1-x}$P$_x$)$_2$ has a higher $T_0$ and $T_c$ than Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$, and these “122” iron-pnictide superconductors have intermediate values of $T_0$ and $T_c$ among other unconventional superconductors. This suggests that the physics of “122” iron-pnictide superconductors may be more closely related to the physics of HF and cuprate superconductors than previously expected, and they may be classified into magnetically mediated superconductors. Moreover, only the optimal superconductivity lies on the curve. This suggests that quantum criticality is another important ingredient for understanding of the linearity between spin fluctuation temperature and $T_c$, as discussed in the HF superconductors such as CeMn$_5$. In addition, it is noteworthy that $T_A$ is roughly scaled to $T_c$ as shown in Fig. 10 (b), and hence to $T_0$. This implies that the spin-fluctuation spectra are renormalized.

FIG. 10: (Color online) The SC transition temperature $T_c$ vs. the spin fluctuation temperature (a) $T_0$ and (b) $T_A$ for various superconductors. The data for $T_c$ and $T_0$ were cited from Refs. 35 and 37. The dotted lines represent linear curve fittings. $T_c$ is linear with $T_0$ as suggested in Ref. 35, and the iron-pnictide superconductors lie on the same line. This may be the signature that Fe pnictides, heavy fermion, and cuprate superconductors are mediated by AF spin fluctuations.
are related with their unconventional superconductivity.

is surprising and suggests that spin-fluctuation spectra
malized by (1.25 $T_c$) are adopted in order to avoid the suppression by the pseudogap effect. The characteristic energy of the spin fluctuations in these compounds seems to be scaled to $T_c$, since the normalized ($T_1 T$)$^{-1}$ data are approximately on the same curve.

C. Coupling between AF spin fluctuations and lattice instability

In this section, we comment on the relationship between magnetism and lattice structure in the “122” compounds.

A structural transition from the high-temperature tetragonal to low-temperature orthorhombic phases occurs at $T_S$ that is identical to $T_N$ or just above $T_N$. Since the structural unit vectors rotates by 45 degree at the transition, the dotted lines in Fig. 12 represent the distorted basal plane below $T_S$, which is a unit cell above $T_S$. An unusual anisotropic interaction ($J_{1a} > J_{1b}, J_{1a} \sim J_2$) was reported in the ordered state from the neutron scattering experiments. We suggest that the anisotropic interactions are reasonably understood by the coupling between four Fe sites by way of the As site as follows. Kita-gawa et al. reported that the electric quadrupole interaction ($\nu$) at the As site changes significantly below $T_S$: $\nu_a$ along the $a$ axis becomes largest, although the difference between the lattice constant $a$ and $b$ is less than 1%. This strongly suggests that the isotropic charge distribution above $T_S$ becomes anisotropic, resulting in a higher electron occupation in $4p_x$ than that in $4p_y$. A similar conclusion was drawn from ARPES experiments. Such an imbalance of occupation implies that degenerate Fe $3d_{xz}$ and $3d_{yz}$ orbitals are lifted due to nonequivalent mixing with As $4p_x$ and $4p_y$ orbitals, in other words, orbital ordering of the Fe $3d$ orbitals is realized. This symmetry breaking naturally leads to a deviation of the exchange interaction $J_1$ between the nearest-neighbor Fe spins. The corresponding orthorhombic distortion can make a $J_{1b}$ ferromagnetic interaction rather than an antiferromagnetic one, following the Goodenough-Kanamori rules, because the Fe-As-Fe bond angle for $J_{1a}$ becomes close to 90 degrees. Such a tendency is consistent with the recent studies by the neutron scattering measurements.

It is naturally expected that the above anisotropic correlations persist well above $T_N$, since the stripe AF fluctuations are observed in the tetragonal phase, and thus the orbital fluctuations linked with the characteristic stripe AF spin correlations are anticipated above $T_N$. Actually, such lattice dynamics was observed with ultrasonic experiments. Goto et al. and Yoshizawa et al. independently that the elastic constant $C_{66}$ in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ shows a large elastic softening towards $T_S$. The latter group pointed out that the Co concentration dependence of the $C_{66}$ softening is ascribable to the presence of a “structural QCP”, similar to a magnetic QCP, and suggested that the high-$T_c$ in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ is related to the structural QCP. Since the temperature dependence of $C_{66}$ is quite similar to that of ($T_1 T$)$^{-1}$ of AF spin fluctuations, we plot the temperature dependence of $C_{66}$ against that of ($T_1 T$)$^{-1}$ with $T$ as an implicit parameter, as shown in Fig. 13. An apparent proportionality between the two quantities strongly suggests that AF spin fluctuations and struc-

FIG. 11: (Color online) The $T$-dependence of ($T_1 T$)$^{-1}$ normalized by ($T_1 T$)$^{-1}$ at 1.25$T_c$ in various unconventional superconductors are plotted against $T/T_c$. ($T_1 T$)$^{-1}$ at 1.25$T_c$ is adopted in order to avoid the suppression by the pseudogap effect. The characteristic energy of the spin fluctuations in these compounds seems to be scaled to $T_c$, since the normalized ($T_1 T$)$^{-1}$ data are approximately on the same curve.

FIG. 12: (Color online) (a) Low-temperature magnetic structure at the Fe layer. The Ba sites are also shown. The dotted lines indicate the tetragonal unit cell above $T_S$, and are deformed below $T_S$. The stripe magnetic structure is shown by the arrows. (b) As $4p_x$,$y$ and Fe $3d_{yz}$ and $3d_{xz}$ orbitals are shown. The difference of the electronic population is shown by contrasting density. Magnetic interactions between nearest neighbor, next nearest neighbor are denoted by arrows.

with $T_0$ as indeed inferred from Fig. 11, where the renormalized ($T_1 T$)$^{-1}$ of various unconventional superconductors approximately scales onto a same curve against $T/T_c$. Because $T_c$ values in these superconductors are different by two orders of magnitude, the scaling of ($T_1 T$)$^{-1}$ is surprising and suggests that spin-fluctuation spectra are related with their unconventional superconductivity.
The iron-pnictide compounds are thus a unique system, where the spin and orbital degrees of freedom are strongly coupled with each other. Although we indicated here that the spin-fluctuation theory is successfully applicable to the 122 systems, the interplay between the spin fluctuation and the orbital degrees of freedom remains to be solved in the future.

V. CONCLUSION

We show that the temperature dependencies of the NMR nuclear spin-lattice relaxation rate, the electrical resistivity, and the inelastic neutron scattering data in the paramagnetic phase of iron-pnictide superconductors BaFe$_2$(As$_{1-x}$P$_x$)$_2$ and Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ can be understood quantitatively in the framework of the SCR theory. A consistent description of these physical properties of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ and Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ in the framework of the SCR theory suggests that an itinerant picture is at work for the “122” iron-pnictide superconductors at the low energy scale and AF quantum criticality would be deeply related to the high-$T_c$ superconductivity, as in other unconventional superconductors. However, a puzzling question in iron-pnictide superconductors is whether AF spin fluctuations and superconductivity are deeply related in “1111” systems such as LaFeAs(O$_{1-x}$F$_x$) and Ca(Fe$_{1-x}$Co$_x$)AsF. In addition, the phase diagram in these systems is different from that in the Ba-“122” systems. Indeed, it was reported recently that superconductivity in LaFeAs(O$_{1-x}$F$_x$) possesses a two-maximum structure and survives until higher hole concentration. It seems that the superconductivity can be observed in the region away from the AF QCP, indicating that the scenario of superconductivity induced by AF spin fluctuations may not be applied universally. Whether a unified picture exists for explaining all experimental results in iron-pnictide superconductors, or whether there exist mechanisms other than magnetism are a future important issue to be clarified.

Acknowledgments

We are grateful to S. Yonezawa and Y. Maeno for fruitful discussions. This work is supported by the Grants-in-Aid for Scientific Research on Innovative Areas “Heavy Electrons” (No. 20102006) from MEXT, for the GCOE Program “The Next Generation of Physics, Spun from Universality and Emergence” from MEXT, and for Scientific Research from JSPS. Y.N. is supported by KAKENHI (No. 23654120).
P. Vilmercati, A. Fedorov, F. Bondino, F. Offi, G. Panaccione, P. L. Ning, K. Ahilan, T. Imai, A. Sefat, M. A. McGuire, B. C. Sales, D. Mandrus, P. Cheng, B. Shen, and H.-H. Wen, Phys. Rev. Lett. 104, 037001 (2010).

Y. Nakai, T. Iye, S. Kitagawa, K. Ishida, H. Ikeda, S. Kasahara, H. Shishido, T. Shibauchi, Y. Matsuda, and T. Terashima, Phys. Rev. Lett. 105, 107003 (2010).

T. Iye, Y. Nakai, S. Kitagawa, K. Ishida, S. Kasahara, T. Shibauchi, Y. Matsuda, and T. Terashima, J. Phys. Soc. Jpn. 81, 033701 (2012).

T. Iye, Y. Nakai, S. Kitagawa, K. Ishida, S. Kasahara, T. Shibauchi, Y. Matsuda, and T. Terashima, Phys. Rev. B 85, 184505 (2012).

J. Dai, Q. Si, J.-X. Zhu, and E. Abrahams, Proc. Natl. Acad. Sci. 106, 4118 (2009).

S. Sachdev and B. Keimer, Physics Today 64, 29 (2011).

Y. Nakai, T. Iye, S. Kitagawa, K. Ishida, S. Kasahara, T. Shibauchi, Y. Matsuda, and T. Terashima, Phys. Rev. B 81, 020503(R) (2010).

K. Hashimoto, M. Yamashita, S. Kasahara, Y. Senshu, N. Nakata, S. Tonegawa, K. Ikada, A. Serafin, A. Carrington, T. Terashima, et al., Phys. Rev. B 81, 220501 (2010).

J. S. Kim, P. J. Hirschfeld, G. R. Stewart, S. Kasahara, T. Shibauchi, T. Terashima, and Y. Matsuda, Phys. Rev. B 81, 214507 (2010).

M. Yamashita, Y. Senshu, T. Shibauchi, S. Kasahara, K. Hashimoto, D. Watanabe, H. Ikeda, T. Terashima, I. Vekhter, A. B. Vorontsov, et al., Phys. Rev. B 84, 060507 (2011).

K. Hashimoto, K. Cho, T. Shibauchi, S. Kasahara, Y. Mizukami, R. Katsumata, Y. Tsuruhara, T. Terashima, H. Ikeda, M. A. Tanatar, et al., Science 336, 1554 (2012).

A. E. Böhm, P. Burger, F. Hardy, T. Wolf, P. Schweiss, R. Fromknecht, H. v. Löhnycsen, C. Meingast, H. K. Mak, R. Lortz, et al., Phys. Rev. B 86, 094521 (2012).

P. Dai, J. Hu, and E. Dagotto, Nature Physics 8, 709 (2012).

H. Gretarsson, A. Lupascu, J. Kim, D. Casa, T. Gog, W. Wu, S. R. Julian, Z. J. Xu, J. S. Wen, G. D. Gu, et al., Phys. Rev. B 84, 100509 (2011).

P. Vilmercati, A. Fedorov, F. Bondino, F. Offi, G. Panaccione, P. L. Ning, L. Simonelli, M. A. McGuire, A. S. M. Sefat, D. Mandrus, et al., Phys. Rev. B 85, 220503 (2012).

L. P. Gor’kov and G. B. Teitel’baum, Phys. Rev. B 87, 024504 (2013).

H. Ikeda, J. Phys. Soc. Jpn. 77, 123707 (2008).

T. Moriya, Y. Takahashi, and K. Ueda, J. Phys. Soc. Jpn. 59, 2905 (1990).

M. Ishikado, Y. Nagai, K. Kodama, R. Kajimoto, M. Nakamura, Y. Inamura, S. Wakimoto, H. Nakamura, M. Machida, K. Suzuki, et al., Phys. Rev. B 84, 144517 (2011).

K. Kitagawa, N. Katayama, K. Ohgushi, and M. Takigawa, J. Phys. Soc. Jpn. 78, 063706 (2009).

S. Kitagawa, Y. Nakai, T. Iye, K. Ishida, Y. Kamihara, M. Hirano, and H. Hosono, Phys. Rev. B 81, 212502 (2010).

K. Kitagawa, N. Katayama, K. Ohgushi, M. Yoshida, and M. Takigawa, J. Phys. Soc. Jpn. 77, 114709 (2008).

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

Y. Nakai, S. Kitagawa, K. Ishida, Y. Kamihara, M. Hirano, and H. Hosono, New J. Phys. 11, 045004 (2009).

K. Matan, S. Ibuka, T. Imai, A. Sefat, M. A. McGuire, B. C. Sales, D. Mandrus, P. Cheng, B. Shen, and H. Wen, arXiv:0910.1071 (2010).

D. S. Inosov, J. T. Park, P. Bourges, D. L. Sun, Y. Sidis, A. Schneidewind, K. Hradil, D. Haug, C. T. Lin, B. Keimer, et al., Nature Physics 6, 178 (2010).

T. Moriya and T. Takimoto, J. Phys. Soc. Jpn. 64, 960 (1995).

S. Kambe, H. Sakai, Y. Tokunaga, T. D. Matsuda, Y. Haga, H. Chudo, and R. E. Walstedt, Phys. Rev. B 77, 134418 (2008).

X. F. Wang, T. Wu, G. Wu, R. H. Liu, H. Chen, Y. L. Xie, and X. H. Chen, New J. Phys. 11, 045003 (2009).

F. Hardy, P. Burger, T. Wolf, R. A. Fisher, P. Schweiss, P. Adelmann, R. Heid, R. Fromknecht, R. Eder, D. Ernst, et al., EuroPhys. Lett. 91, 47008 (2010).

T. Moriya and K. Ueda, J. Phys. Soc. Jpn. 63, 1871 (1994).

T. Moriya and K. Ueda, Rep. Prog. Phys. 66, 1299 (2003).

T. Moriya and K. Ueda, Adv. Phys. 49, 555 (2000).

C. Pfleiderer, Rev. Mod. Phys. 81, 1551 (2009).

N. J. Curro, T. Caldwell, E. D. Bauer, L. A. Morales, M. J. Graf, Y. Bang, A. V. Balatsky, J. D. Thompson, and J. L. Sarrao, Nature 434, 622 (2005).

P. Gegenwart, Q. Si, and F. Steglich, Nature Phys. 4, 186 (2008).

T. Park, F. Ronning, H. Q. Yuan, M. B. Salamon, R. Movshovich, J. L. Sarrao, and J. D. Thompson, Nature 440, 65 (2006).

K. Ishida, Y. Kitaoka, N. Ogata, T. Kamino, K. Asayama, J. R. Cooper, and N. Athanassopoulos, J. Phys. Soc. Jpn. 62, 2803 (1993).

S. Ohsumi, Y. Kitaoka, K. Ishida, and K. Asayama, J. Phys. Soc. Jpn. 60, 2351 (1991).

Y. Kawasaki, S. Kawasaki, M. Yashima, T. Mito, G. q. Zheng, Y. Kitaoka, H. Shishido, R. Settai, Y. Haga, and Y. Onuki, J. Phys. Soc. Jpn. 72, 2308 (2003).

K. Ishida, Y. Kawasaki, K. Tabuchi, K. Kashiwa, Y. Kitaoka, K. Asayama, C. Geibel, and F. Steglich, Phys. Rev. Lett. 82, 3533 (1999).

D. C. Johnston, Advances in Physics 59, 803 (2010).

T. Shimojima, K. Ishizaka, Y. Ishida, N. Katayama, K. Ohgushi, T. Kiss, M. Okawa, T. Togashi, X.-Y. Wang,
C.-T. Chen, et al., Phys. Rev. Lett. 104, 057002 (2010).

50 M. Yoshizawa, D. Kimura, T. Chiba, S. Simayi, Y. Nakanishi, K. Kihou, C. H. Lee, A. Iyo, H. Eisaki, M. Nakajima, et al., J. Phys. Soc. Jpn. 81, 024604 (2012).

51 T. Goto, R. Kurihara, K. Araki, K. Mitsumoto, M. Akatsu, Y. Nemoto, S. Tatematsu, and M. Sato, J. Phys. Soc. Jpn. 80, 073702 (2011).

52 S. Kasahara, H. J. Shi, K. Hashimoto, S. Tonegawa, Y. Mizukami, T. Shibauchi, K. Sugimoto, T. Fukuda, T. Terashima, A. H. Nevidomskyy, et al., Nature 486, 382 (2012).

53 Y. Kobayashi, E. Satomi, S. C. Lee, and M. Sato, J. Phys. Soc. Jpn. 79, 093709 (2010).

54 T. Oka, Z. Li, S. Kawasaki, G. F. Chen, N. L. Wang, and G.-q. Zheng, Phys. Rev. Lett. 108, 047001 (2012).

55 S. Tsutsumi, N. Fujiwara, S. Matsuishi, and H. Hosono, Phys. Rev. B 86, 060515 (2012).

56 S. Iimura, S. Matsuishi, H. Sato, T. Hanna, Y. Muraba, S. W. Kim, J. E. Kim, M. Takata, and H. Hosono, Nature Communications 3, 943 (2012).