P-concentration dependence of the quasiparticle density of states in BaFe$_2$(As$_{1-x}$P$_x$)$_2$

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Abstract. $^{31}$P-NMR measurements were performed in the iron pnictides BaFe$_2$(As$_{1-x}$P$_x$)$_2$ for $0.07 \leq x \leq 0.64$ in order to investigate evolution of electronic state with isovalent substitution. In general, the spin part of the Knight shift $K_{\text{spin}}$ and $(T_cT)^{-1/2}$ are proportional to the quasiparticle density of states at the Fermi level $N(E_F)$ at high temperature where the Korringa relation holds. We found that $K_{\text{spin}}$ and $(T_cT)^{-1/2}$ are almost unaffected by the P-substitution, indicating that $N(E_F)$ is nearly constant. These results are quantitatively consistent with our band calculation. We claim that the slight changes of $N(E_F)$ by P-substitution in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ is not enough to explain the $T_c$ variation ranging from 30 K to 0 K, and thus suggest that antiferromagnetic correlations related to the quantum critical character are essential for the high $T_c$ superconductivity.

Despite a large number of experimental and theoretical researches on the physics of recently discovered iron-pnictides [1], the mechanism of high-$T_c$ superconductivity is still under debate. Spin-fluctuation-mediated superconductivity which promotes the $s_\pm$-wave superconducting gap symmetry with a sign change between electron and hole bands is one of the most promising scenario [2, 3, 4, 5, 6], while orbital-fluctuation-mediated superconductivity whose gap symmetry is $s_{++}$ with no sign change is considered to be another candidate [7]. In case of conventional phonon-mediated superconductivity, $T_c$ is closely related to the quasiparticle density of states at the Fermi level $N(E_F)$ in the form of $T_c = 1.14 \omega_0 e^{-2/gN(E_F)}$, where $\omega_0$ is the characteristic frequency of collective excitations involved in pairing interaction and $g$ is the appropriate dimensional coupling constant. Since there is a possibility that the changes in $T_c$ by chemical substitution and/or physical pressure is attributed to the changes in $N(E_F)$, it should be examined with a proper system to discuss the nature of dominant pairing interaction.

In this sense, we studied BaFe$_2$(As$_{1-x}$P$_x$)$_2$ as such an ideal system. This is because the substitution of isovalent P for As is expected to introduce chemical pressure without doping charge carriers in contrast with hole-doping (Ba$_{1-x}$K$_x$)Fe$_2$As$_2$ and electron-doping Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$. BaFe$_2$(As$_{1-x}$P$_x$)$_2$ shows unconventional superconductivity at $T_c^{\text{max}} = 31$ K ($x \sim 0.30$) [8, 9, 10, 11, 12] with line nodes in the superconducting gap [13, 14, 15, 17, 16, 18]. However, recent ARPES experiment revealed that P-substitution in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ induced significant hole-doping [19]. This result suggests that the iso-valent picture in BaFe$_2$(As$_{1-x}$P$_x$)$_2$
is oversimplified [19], and hence, it is important to investigate the variation of $N(E_F)$ experimentally. Our investigation clarified that $N(E_F)$ remains almost unchanged by P-substitution, while the characteristic temperature of magnetic anomaly $\theta$ deduced from $(T_1 T)^{-1}$ largely changes with a sign change across an anticipated quantum critical point. These suggest that the change in antiferromagnetic correlations related to the quantum critical character is more important than the change in $N(E_F)$ for the $T_c$ variation in the P-substituted system.

Samples of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ for $0.07 \leq x \leq 0.64$, consisting of single crystals with average dimensions of $100 \times 100 \times 50 \mu m^3$ were prepared as described elsewhere [9]. $^{31}$P-NMR spectra were obtained by sweeping frequency in a fixed magnetic field of 4.12 T at 200 K. The Knight shift $K$ was determined from the peak frequency with respect to the reference material H$_3$PO$_4$ ($K = 0$). The $^{31}$P nuclear spin-lattice relaxation rate $T_1^{-1}$ was determined by fitting the time dependence of spin-echo intensity after saturation of nuclear magnetization to a theoretical $I = 1/2$ curve with a single component of $T_1$.

The resonance frequencies of $^{31}$P-NMR spectra consisting of a single line for $0.07 \leq x \leq 0.64$ are temperature-independent in the normal state [10]. The temperature variation of $K$ is shown in Fig.1(a). $K$ in the normal state slightly decrease with increasing P-concentration $x$. In general, $K$ is composed of the temperature-dependent spin shift $K_{spin}$ and the temperature-independent chemical shift $K_{chem}$: $K(T) = K_{spin}(T) + K_{chem}$. $K_{spin}$ is related to the uniform spin susceptibility $\chi(q = 0)$ which is proportional to $N(E_F)$, while $K_{chem}$ is unrelated to $N(E_F)$. Since no obvious AFM fluctuations were detected via $(T_1 T)^{-1}$ at high temperatures as seen in Fig.1(b), it would be a good approximation to consider that $(T_1 T)^{-1/2}$ is proportional to $N(E_F)$ at high temperatures, i.e., to assume that the usual Korringa relation holds. Based on the plot of $(T_1 T)^{-1/2}$ against $K$ at 270 K for different $x$ which is shown in the inset of Fig.1(a), we estimated $K_{chem} = 0.018 \pm 0.019 \%$ from the intercept [10]. This value would be reasonable since $K_{chem}$ for $^{31}$P in many diamagnetic insulators is of the order of some hundreds of ppm [21]. Using the obtained $K_{chem}$ with an assumption of $x$-independent $K_{chem}$, we deduced $K_{spin}$ at 200 K for each $x$ and plotted them in Fig.2(a). Since $K_{spin}$ is proportional to $N(E_F)$, the

![Figure 1](Color online) (a) The temperature dependences of Knight shifts for $0.07 \leq x \leq 0.64$ in BaFe$_2$(As$_{1-x}$P$_x$)$_2$. The black arrow denotes the value of $K_{chem} = 0.018 \pm 0.019 \%$ determined by $(T_1 T)^{-1/2} - K$ plot (inset). (b) The temperature dependences of $(T_1 T)^{-1}$ for $0.07 \leq x \leq 0.64$ in BaFe$_2$(As$_{1-x}$P$_x$)$_2$. The solid lines are the fitting curves to $(T_1 T)^{-1} = a + b(T + \theta)^{-1}$. The inset shows $x$-dependences of the fitting parameters $a$ and $\theta$, and the dotted line corresponds to 0 K in the right axis.
dependence of $K_{\text{spin}}$ suggests that the change in $N(E_F)$ is at most 16 \% for superconducting $0.20 \leq x \leq 0.64$. This result is quantitatively consistent with the change in $N(E_F)$ deduced from $(T_1T)^{-1/2}$ and our band calculation. The calculations by local-density-approximation were performed for nonspin-polarized BaFe$_2$As$_2$ and BaFe$_2$P$_2$, using the WIEN2K package in the APW + local orbital basis [10]. We performed the local-density-approximation calculations for BaFe$_2$As$_2$ with linearly-interpolated $(a, c, z) = [a_0(1-x) + a_1x, c_0(1-x) + c_1x, z_0(1-x) + z_1x]$, where $a_0(1)$, $c_0(1)$, and $z_0(1)$ are the experimental values for the crystallographic parameters of BaFe$_2$As$_2$(BaFe$_2$P$_2$) [9, 26]. The calculated $N(E_F)$ are plotted as a solid line in Fig.2(a), which agree with the experimental results quantitatively.

Low energy spin fluctuations are probed by measuring $T_1^{-1}$. AFM spin fluctuations are enhanced significantly with decreasing $x$ from $x = 0.64$ where $(T_1T)^{-1}$ is almost constant. The measured $(T_1T)^{-1}$ can be well fit by the equation expected from the self-consistent renormalization (SCR) theory under the existence of two-dimensional (2D) AFM spin fluctuations [22]: $(T_1T)^{-1} = a + b(T + \theta)^{-1}$, which are shown as solid lines in Fig.1(b). The first term is the contribution originating from intraband scattering and is proportional to $N(E_F)^2$, the second term is the contribution of interband AFM spin fluctuation. $x$-dependence of $a$ and $\theta$ are demonstrated in the inset of Fig.1(b). With increasing $x$, the parameter $a$ decreases slightly whereas $\theta$ increases significantly with a sign change around $x \sim 0.33$ where quantum critical point is anticipated.

We plot the $x$-dependence of $(T_1T)^{-1/2}$ at high temperatures for BaFe$_2$(As$_{1-x}$P$_x$)$_2$, (Ba$_{1-x}$K$_x$)Fe$_2$As$_2$ [23], and Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [24] to compare the evolution of $N(E_F)$. The K- and Co-substitution induce significant changes in $N(E_F)$, while the P-substitution has little effects on $N(E_F)$. Note that the changes in $N(E_F)$ for superconducting dome region $0.2 \leq x \leq 1$ in (Ba$_{1-x}$K$_x$)Fe$_2$As$_2$ are ~60\% increase and $0.04 \leq x \leq 0.14$ in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ are ~20\% decrease from the value of BaFe$_2$As$_2$, whereas the change in BaFe$_2$(As$_{1-x}$P$_x$)$_2$ is only 11 \% decrease. These results suggest that the $N(E_F)$ change is not a primary role for superconductivity in the “122” compounds. Rather, the AFM fluctuations with the quantum critical character are more important for superconductivity than the $N(E_F)$ change.

Figure 2. (Color online) (a) $x$-dependence of $K_{\text{spin}}$ and $^{31}$P ($T_1T)^{-1/2}$ at 270 K of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ and $x$-evolution of $N(E_F)$ derived from the band calculation. (b) $x$-dependence of $^{31}$P ($T_1T)^{-1/2}$ at 270 K of BaFe$_2$(As$_{1-x}$P$_x$)$_2$, $^{75}$As ($T_1T)^{-1/2}$ at 280 K of (Ba$_{1-x}$K$_x$)Fe$_2$As$_2$ [23] and Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [24], all of which are proportional to $N(E_F)$. The dotted lines are guides to the eye. The vertical axis of $^{31}$P ($T_1T)^{-1/2}$ is adjusted to unify the value of BaFe$_2$As$_2$ with that in the right axis.
In summary, we have investigated the P-substitution effect to the electronic state in BaFe$_2$As$_2$ through $^{31}$P-NMR. The quasiparticle density of states $N(E_F)$ detected via measuring the spin part of the Knight shifts $K_{\text{spin}}$ and the nuclear spin-lattice relaxation rate $T_1^{-1}$ show only slight decrease with increasing P-concentration $x$, quantitatively consistent with the results of the band calculation. Almost unaffected value of $N(E_F)$ by P-substitution is compared to more rapid changes in $N(E_F)$ caused by K and Co doping. Our experiments clarified that $T_c$ in the “122” system is not mainly determined by the $N(E_F)$ change but by the AFM spin fluctuations.

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