NMR study under pressure on the La1111 pnictides, $LaFeAsO_{1-x}F_x$

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Abstract

The relationship between antiferromagnetic (AF) fluctuation and superconductivity was investigated in the La1111 series, $LaFeAsO_{1-x}F_x$ (x = 0.05, 0.08, and 0.14) by examining nuclear relaxation rates ($1/T_1$) at both ambient pressure and 3.0 GPa. Although the AF fluctuation is enhanced by applying pressure in the underdoped regime ($x = 0.05$, and 0.08), the increase in critical transition temperature (T_c) is small, whereas T_c increases remarkably in the overdoped regime ($x = 0.14$) where the AF fluctuation is absent, suggesting that high T_c above 40 K originates not from the AF fluctuation but from the density of states at the electron pocket.

KEYWORDS NMR, High pressure, La 1111 series

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I. INTRODUCTION

The 1111 series ($RFeAsO_{1-x}F_x$ R=Nd, Sm, Ce, and La, *etc.*) is highly important because its critical transition temperature (T_c) is relatively high among iron-based pnictides [1-7]. In the 1111 series, the optimal T_c is realized away from an antiferromagnetic (AF) phase on the $T - x$ phase diagram, and the superconductivity is maintained even in a heavily doped regime. These features contrast with those of the 122 series, in which the optimal T_c appears adjacent to the AF phase [8-10].

In the La1111 series, the optimal T_c of 26 K appears around $x = 0.11$, away from the AF phase. The SC phase is not so sensitive to doping level, but is sensitive to pressure. A clear dome-shaped pressure (P) dependence of T_c has been observed by the resistivity measurements [11, 12]. The highest T_c is realized by applying pressure to optimally doped ($x \sim 0.11$) or heavily doped ($x \sim 0.14$) samples: T_c values, 26 and 20 K for $x = 0.11$ and 0.14, respectively reach to 43 K at a pressure of 4-5 GPa [11, 12]. Structurally, the highest T_c has been realized when Fe and As ions form regular tetrahedra or the pnictogen height above the basal plane of iron is high [13, 14]. Changes in these parameters resulting from pressure application can modify the band structure and covalency between Fe and As ions, affecting the electronic and/or magnetic properties.

However, the origin of high T_c in iron-based pnictides is unclear; spin fluctuation seems to be an improtant factor from an analogy with high-T_c cuprates, while an SC mechanism due to orbital fluctuation has also been suggested [15, 16]. To investigate whether the superconductivity is of magnetic origin as it is in high-T_c cuprates, we measured nuclear magnetic relaxation rates ($1/T_1$) of ^{75}As systematically under pressure for several doping levels ($x = 0.05$, 0.08 and 0.14).

II. EXPERIMENTAL CONDITIONS

Powder samples were used for the nuclear magnetic resonance (NMR) measurements of ^{75}As. Field-swept NMR spectra were measured at a fixed frequency of 45.1 MHz. The spectra exhibit a typical powder pattern with two peaks, which is seen under nuclear quadrupole interactions. The relaxation rate $1/T_1$ was measured at the lower-field peak by the saturation-recovery method. The Fe-As planes contributing to this peak get aligned parallel to the

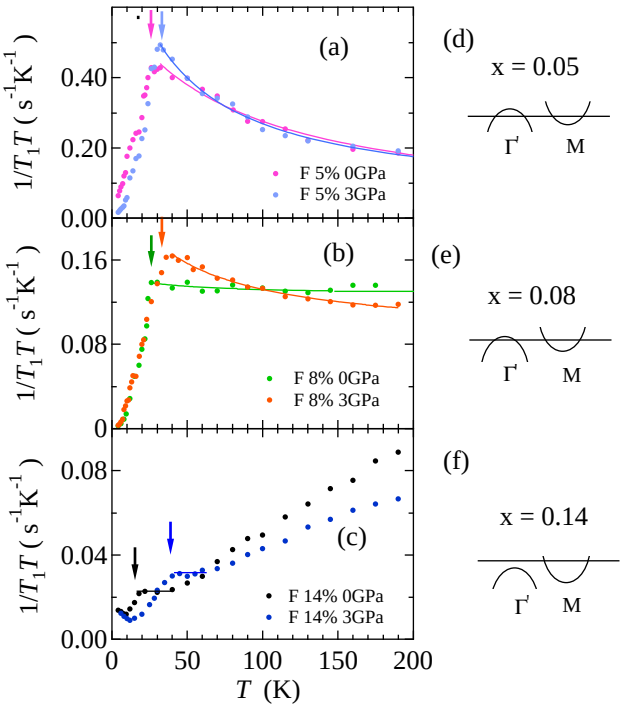

FIG. 1: Relationship between antiferromagnetic (AF) fluctuation and band structure. (a-c) Temperature dependence of $1/T_1T$ at several doping levels. Arrows indicate T_c determined from changes in $1/T_1T$. (d-f) Schemes of electron and hole pockets. The Γ' point represents (π, π) in the unfolded Brilliouin Zone. That point overlaps the Γ point corresponding to $(0, 0)$ in the original folded Brilliouin Zone.

applied field.

III. EXPERIMENTAL RESULTS

Figs. 1(a)-(c) show $1/T_1T$ measured at ambient pressure and 3.0 GPa for the samples doped with x = 0.05, 0.08 and 0.14. The data for x= 0.08 and 0.14 were cited from Ref. [17,18]. For the x = 0.14 samples, $1/T_1T$ has been measured up to 3.7 GPa [18]. At each doping level, qualitatively different features are revealed by applying pressure. Below, we describe in detail what occurs at each doping level.

1) Lightly doped regime (x = 0.05). Samples with a very low doping level exhibit clear Curie-Weiss behavior, suggesting that low-frequency AF fluctuation is predominant at this

doping level. The Curie-Weiss behavior is enhanced and T_c increases slightly at a pressure of 3.0 GPa. The increase ΔT_c, indicated by arrows in Fig. 1(a), is estimated as 7-8 K.

2) Underdoped regime ($x = 0.08$). For the $x = 0.08$ samples, Curie-Weiss behavior is weaker than for $x = 0.05$ and appears almost independent of T at ambient pressure. However, Curie-Weiss behavior returns when pressure is applied. The increase in T_c is almost the same as for $x = 0.05$.

3) Over doped regime ($x = 0.14$). For the $x = 0.14$ samples, the Curie-Weiss behavior completely vanishes at both ambient pressure and 3.0 GPa. $1/T_1T$ shows a plateau just above T_c and a monotonous increase with the increase in temperature. Remarkable T_c enhancement is seen by applying a pressure of 3.0 GPa. Pressure application enhances $1/T_1T$ just above T_c, whereas it suppresses $1/T_1T$ at high temperatures. The behavior at temperatures above T_c originates not from the AF fluctuation but from peculiarities of the band structure as described in the following section.

IV. BAND STRUCTURE

The qualitatively different T- and P-dependence of $1/T_1T$ can be explained by the band calculations [19, 20]. According to the band calculations, the AF fluctuation originates from the nesting between hole and electron pockets in the lightly doped regime, as illustrated in Fig. 1(d). In the underdoped regime ($x = 0.08$), the weak Curie-Weiss behavior arises from weak nesting originating from an imbalance between hole and electron pockets. (see Fig. 1(e).) With increasing doping level, γ Fermi surface, a hole pocket near the $\Gamma'(\pi, \pi)$ point in the unfolded Brilliouin zone, vanishes around $x = 0.10$ [14]. However, states near the $\Gamma'(\pi, \pi)$ point just below the Fermi level contribute to the density of states $D(\varepsilon_F)$ at high temperature. The T-dependent $D(\varepsilon_F)$ gives rise to the monotonous increase in $1/T_1T$ ($\sim D(\varepsilon_F)^2$). The band-structure effect disappears at low temperatures, and other hole pockets, α_1 and α_2 surfaces around the $\Gamma(0, 0)$ point, are expected to become small [21]. Therefore, a plateau in $1/T_1T$ just above T_c originates from $D(\varepsilon_F)$ at the electron pocket, β surface around the M point. The remarkable T_c enhancement due to pressure application is attributable to an increase in $D(\varepsilon_F)$ at the β surface. Based on the band calculations, the $1/T_1T$ components coming from T-dependent and T-independent $D(\varepsilon_F)$ have different origins. This could explain why $1/T_1T$ exhibits a qualitatively different pressure response

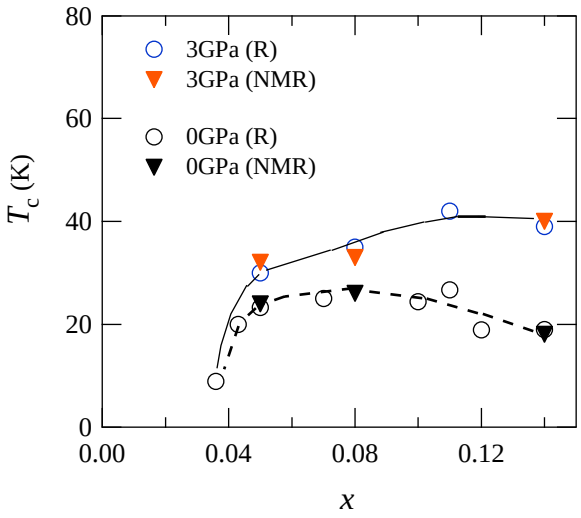

FIG. 2: T_c determined by the resistivity and $1/T_1T$ at ambient pressure and 3 GPa. The values of T_c shown by arrows in Figs. 1(a)-(c) are plotted by triangles.

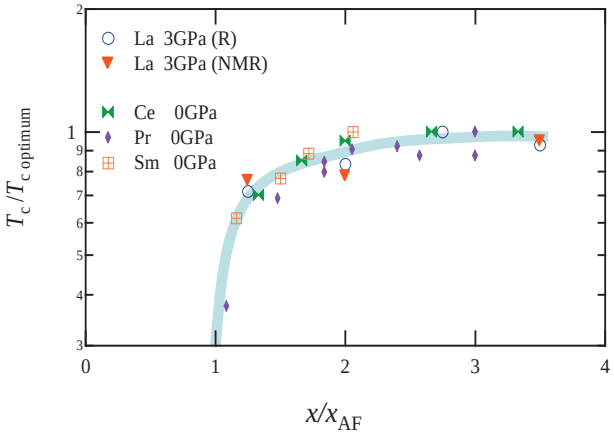

FIG. 3: T_c for various $RFeAsO_{1-x}F_x$. T_c and x are normalized by the optimal T_c and the antiferromagnetic phase boundary x_{AF}, respectively. Blue circles and red triangles represent T_c determined from the resistivity and $1/T_1T$ at 3 GPa, respectively.

between low and high temperatures, as shown in Fig. 1(c): $1/T_1T$ at high temperature is suppressed by applying pressure, whereas the plateau in $1/T_1T$ is enhanced by applying pressure.

V. DISCUSSION

The values of T_c determined from the onset of the resistivity and $1/T_1T$ (arrows in Figs. 1(a)-(c)) are plotted in Fig. 2. Pressure application enhances low-frequency AF fluctuation in the underdoped regime. However, the increase in T_c is small. As the figure shows, superconductivity with $T_c \geq 40K$ develops in the overdoped regime away from the strong AF fluctuation caused by pressure application. This fact calls into question the strong interplay between AF fluctuation and superconductivity. Thus, the question of whether the features observed in the overdoped regime are common to the other 1111 series arises.

The $T - x$ phase diagram at 3.0 GPa is reminiscent of diagrams of the Ce, Pr and Sm 1111 series at ambient pressure in that T_c hardly drops to below 40 K even in the heavily doped regime, and the highest T_c is realized away from the AF phase. Fig. 3 shows a phase diagram normalized by the doping levels, x_{AF}, at which the AF phase vanishes. The values of x_{AF} are estimated as 0.04, 0.06, 0.075, and 0.04 for the La, Ce, Pr, and Sm 1111 series, respectively [1-7]. The phase diagram includes some ambiguity in the determination of x_{AF}. However, Fig. 3 allows comparison of the SC phase boundary because differences due to x-estimation methods are excluded to some extent. As the figure shows, the x/x_{AF} dependence of T_c normalized by the optimal T_c is almost the same for the 1111 series with high T_c above 40 K.

The similarity between the La1111 series at 3.0 GPa and the other 1111 series is well understood if the pnictogen height from the basal plane of iron determines T_c, as suggested by a theoretical investigation [14]. According to x-ray diffraction measurements under pressure by Garbarino, *et. al.*, pnictogen height increases with increasing pressure, and the lattice constant shrinks as well [22]. The same changes occur by complete replacement of rare-earth ions: the pnictogen height increases in the order of La, Ce, Nd, and Sm, and the lattice constants also shrink in the same order [3, 6, 23-25]. The La1111 series at 3.0 GPa ($T_c = 40$ K) corresponds to the Ce1111 series at ambient pressure. The pnictogen height and lattice parameter of the Ce1111 series are 0.1565 and 3.97 $\mathring{A}$, respectively [6]. According to the x-ray diffraction measurements on the La 1111 series, the former and latter are estimated to be 0.158 and 3.97 $\mathring{A}$, respectively, at 3.16 GPa [22]. The agreement is fairly good, therefore, the La1111 series at 3.0 GPa and the Ce1111 series at ambient pressure are equivalent, and the phase diagram determined under pressure is common to the 1111 series with high

T_c above 40 K. In some respects, pressure application to the La1111 series is very useful to investigate the high-T_c mechanism in the other 1111 series with $T_c \geq 40$ K, because in the other 1111 series magnetic fluctuation arising from rare-earth ions predominates, which prevents the extraction of AF fluctuation arising from the basal planes of iron.

VI. SUMMARY

In summary, the relationship between the AF fluctuation and superconductivity was investigated by using NMR under high pressure, suggesting that high T_c above 40 K in the 1111 series originates not from the AF fluctuation but from $D(\varepsilon_F)$ at the electron pocket around the M point.

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