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Experimental evidence of T_c enhancement without the influence of spin fluctuations: NMR study on LaFeAsO$_{1-x}$H$_x$ under a pressure of 3.0 GPa

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The electron-doped high-transition-temperature (T_c) iron-based pnictide superconductor LaFeAsO$_{1-x}$H$_x$ has a unique phase diagram: Superconducting double domes are sandwiched by antiferromagnetic phases at ambient pressure and they turn into a single dome with a maximum T_c that exceeds 45 K at a pressure of 3.0 GPa. We studied whether spin fluctuations are involved in increasing T_c under a pressure of 3.0 GPa by using the ^{75}As nuclear magnetic resonance (NMR) technique. The ^{75}As-NMR results for the powder samples show that T_c increases up to 48 K without the influence of spin fluctuations. This fact indicates that spin fluctuations are not involved in raising T_c, which implies that other factors, such as orbital degrees of freedom, may be important for achieving a high T_c of almost 50 K.

The phase diagram of the electron-doped high-transition-temperature (T_c) iron-based pnictide LaFeAsO$_{1-x}$H$_x$ (H-doped La1111 series) is unique owing to the capability of electron doping: (i) It exhibits a superconducting (SC) phase with double domes covering a wide H-doping range from $x = 0.05$ to 0.44 [1], (ii) the SC phase is sandwiched by antiferromagnetic (AF) phases appearing in heavily and poorly electron-doped regimes [see Fig. 1(a)] [2], and (iii) the application of pressure transforms the double domes into a single dome [1,3]. Intriguingly, upon applying pressure, the minimum T_c at ambient pressure becomes the maximum T_c of over 45 K [1], as shown by the solid arrow in Figs. 1(a) and 4, as described in detail below.

The unique features established in this compound have not been observed so far in other iron-based pnictides, such as the Ba122 and Na111 series, which have been investigated intensively from an early stage because they are available as large crystals. The electronic phase diagram of the Ba122 series is similar to that of high-T_c cuprates [4]. The analogy is reminiscent of the importance of AF fluctuations in iron-based pnictides. The spin-fluctuation-mediated mechanism is a major candidate for the high-T_c mechanism. In fact, the SC phase of the Ba122 series partially overlaps the AF phase, in other words, the SC and AF states are compatible, and the maximum T_c occurs close to the phase boundary [see Fig. 1(b)] [5,6]. Because of this special location, T_c is enhanced and low-energy AF fluctuations simultaneously become predominant as the doping level approaches the AF phase [7,8]. The Na111 series has a phase diagram similar to that of the Ba122 series, however, the SC phase overlaps the AF phase over a wide doping range and even extends to the undoped material [9]. By tuning pressure, T_c and AF fluctuations are found to be related in a similar manner as in the Ba122 series [10]. A pressure-enhanced T_c occurs in the 11 series FeSe, which is superconducting and has no magnetic orders at ambient pressure. At first sight, the series seems to be free from antiferromagnetism, however, at pressures exceeding 1 GPa,

the SC phase is adjacent to the AF or AF+SC phase [11]. In fact, the influence of AF fluctuations is observable in the SC phase even at ambient pressure [12].

AF fluctuations seem to play a key role in raising T_c for various iron-based pnictides, however, the scenario does not work well for LaFeAsO$_{1-x}$F$_x$, because, for $x = 0.14$, T_c increases up to 40 K at 3.0 GPa with no predominant AF fluctuations [13,14]. The La1111 series under high pressure is the only material available for investigating the magnetic properties of pnictides with T_c in the range of 45–50 K. In fact, the Sm1111 series marks the highest T_c ($T_c = 55$ K) in all types of iron-based pnictides [15], however, it includes magnetic Sm ions, which hinders the investigation of the magnetic properties of iron-basal planes. The rise of T_c without AF fluctuations was observed only for $x = 0.14$ (see the dashed arrow in Fig. 4), because AF fluctuations remain in a lower doping range than $x = 0.14$ and, unfortunately, $x = 0.14$ is nearly the highest level of F doping. As far as nuclear magnetic resonance (NMR) studies on LaFeAsO$_{1-x}$F$_x$ are concerned, the maximum doping level is less than $x = 0.14$–0.15 [16,17]. To establish the breakdown of this scenario over a wide doping range ($0.20 \leqslant x \leqslant 0.44$) that covers the second SC dome, we applied ^{75}As ($I = 3/2$) NMR to powder samples of the H-doped La1111 series at 3.0 GPa.

We applied a pressure of 3.0 GPa to samples with $x = 0.20$, 0.40, and 0.44. The pressure was applied by using NiCrAl-CuBe hybrid piston-cylinder-type cells. We used a mixture of F-70 and F-77 fluorinate as the pressure mediation liquid. The details of the pressure cells are given in Ref. [18]. A coil wound around the samples inside the pressure cell and capacitors equipped with a NMR probe form the tank circuit, which serves to detect the detuning of the resonance frequency, namely, the ac susceptibility, and to detect the NMR signal as well. The NMR measurements were performed using a conventional coherent pulsed-NMR spectrometer. The ^{75}As-NMR spectra show a broad powder pattern with double edges [14], which originates from the second-order quadrupole effect under a magnetic field. The relaxation time (T_1) for ^{75}As was measured by using the saturation-recovery method at the lower-field edge in the field-swept NMR spectra. The low-field

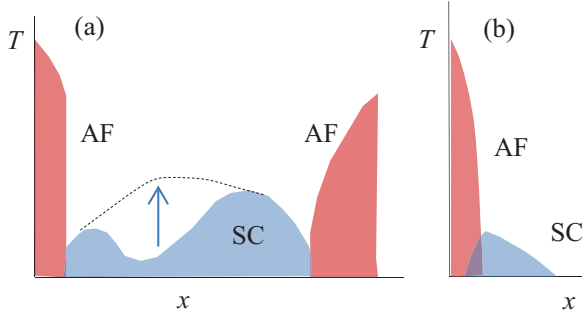———————
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FIG. 1. Schematic phase diagrams of (a) H-doped La1111 series LaFeAsO$_{1-x}$H$_x$, and of (b) Ba122 series such as Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$. AF and SC represent antiferromagnetic and superconducting phases, respectively. The arrow in (a) shows the enhancement of T_c that occurs between ambient pressure and 3.0 GPa.

FIG. 3. Detuning of resonance frequency for (a) $x = 0.20$, (b) $x = 0.40$, and (c) $x = 0.44$. T_c's determined from $1/T_1T$ [see arrows in Figs. 1(a)–1(c)] are in good agreement with the extrapolations of dashed lines.

edge appears at about 48.2 kOe for an NMR frequency of 35.1 MHz. The signals at the low-field edge come from the powder samples with the iron-basal planes parallel to the applied field. Figures 2(a)–2(c) show the evolution of the relaxation rate $(1/T_1)$ divided by temperature (T) (i.e., $1/T_1T$). Here, we chose T_c as the onset of $1/T_1T$, plotted as arrows in Figs. 2(a)–2(c). For $x = 0.20$, $1/T_1T$ just above T_c exhibited plateaus, as shown by dotted lines in Fig. 2(a), and T_c was remarkably enhanced upon applying pressure. For $x = 0.40$ and 0.44, an anomaly was observed in a paramagnetic state, marked as T^*, and both T^* and T_c were unchanged upon applying pressure. For $x = 0.44$, Curie-Weiss-like behavior, which implies AF fluctuations, appears above T^* at 0.1 MPa, however, this behavior has no appreciable effect on T_c.

Note that T_c was determined under the applied field. In general, T_c decreases more or less under an applied field, however, the decrease is significantly suppressed because we measured $1/T_1T$ for the powder samples with the iron-basal planes parallel to the applied field. In fact, the values of T_c are in good agreement with those determined from the detuning of
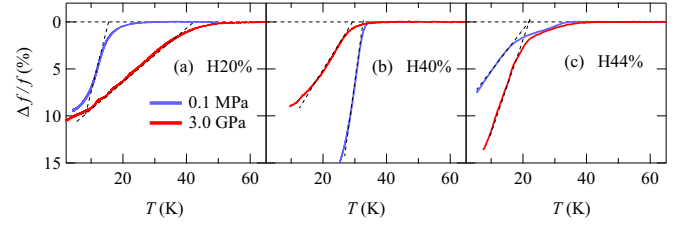
resonance frequency at zero field. In this measurement, T_c can be determined from the extrapolation, shown as dashed lines in Figs. 3(a)–3(c). Figure 4 shows the doping dependence of T_c determined from the resistivity [1] and $1/T_1T$. The data at 3.0 GPa for the low-doping regime are cited from the results of the F-doped La1111 series [14,19]. As highlighted by the solid arrow in Fig. 4, T_c increases to 48 K at 3.0 GPa, which is comparable to the highest T_c ($\sim$ 55 K) for all types of iron-based superconductors marked in the Sm1111 series.

In general, $1/T_1T$ of d-electron systems is determined by spin correlations and is expressed by using the imaginary part of the dynamical spin susceptibility Im $\chi(q,\omega)$ as $1/T_1T \propto$ Im $\chi(q,\omega_N)/\omega_N$, where ω_N is the angular frequency of nuclei. When the interaction between electrons is significantly strong, namely, spin fluctuations are predominant, Curie-Weiss-like behavior is derived, whereas when the interaction is weak, $1/T_1T$ is determined by the density of states (DOS) at Fermi surfaces. Unlike other pnictides, Curie-Weiss-like behavior is not observable for $x = 0.20$, as seen in Fig. 2(a). Another example is K$_y$Fe$_{2-x}$Se$_2$ with $T_c = 30$ K [20]: The compound exhibits a similar T dependence to the La1111 series. The results in Fig. 2(a) demonstrate that T_c is enhanced without appreciable low-frequency AF fluctuations, which is the most important result for this study. The absence of AF fluctuations
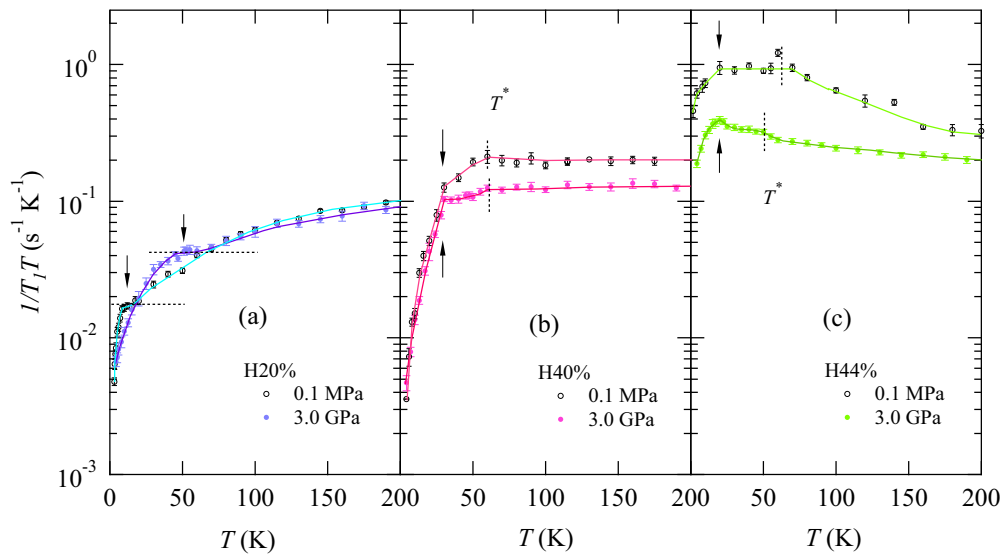

FIG. 2. Nuclear magnetic relaxation rate divided by temperature $1/T_1T$ for ^{75}As for (a) $x = 0.20$, (b) $x = 0.40$, and (c) $x = 0.44$. Arrows represent T_c and solid curves are guides to the eye. Horizontal dotted lines in (a) represent plateaus just above T_c. For $x = 0.40$ and 0.44, an anomaly of $1/T_1T$ appears at T^* in a paramagnetic phase (see vertical dotted lines).
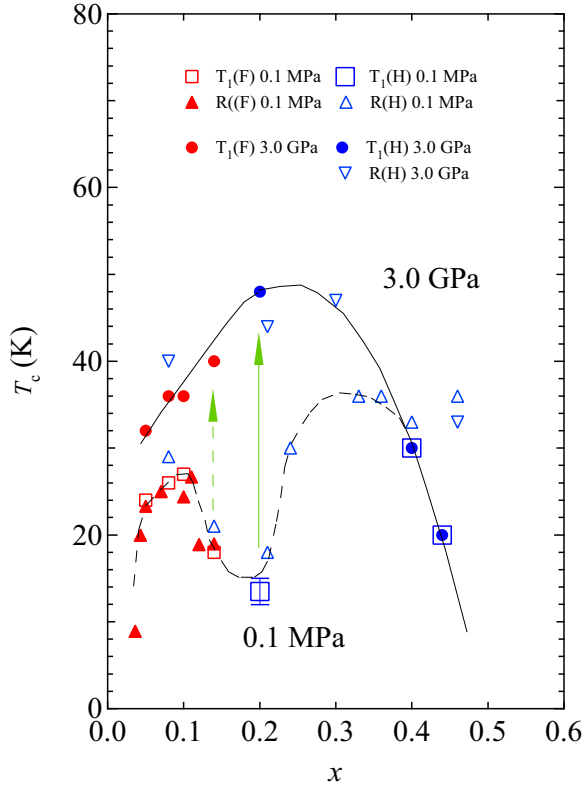
FIG. 4. Phase diagram of LaFeAsO$_{1-x}$H$_x$ (0.05 $\leqslant x \leqslant$ 0.5) and LaFeAsO$_{1-x}$F$_x$ (0.05 $\leqslant x \leqslant$ 0.14). Solid and open triangles represent T_c determined by the resistivity at ambient pressure, and open squares represent T_c determined by the relaxation time (T_1). Downward pointing triangles and solid circles represent T_c determined by the resistivity and T_1 at 3.0 GPa, respectively. The dashed and solid arrows indicate the enhancement of T_c that occurs when pressure is applied.

has also been confirmed from the neutron scattering measurements in both LaFeAsO$_{1-x}$H$_x$ [21] and LaFeAsO$_{1-x}$F$_x$ [22]: An inelastic scattering peak is absent for $x = 0.20$, despite the fact that it is unambiguously observable near the AF phase.

Herein, the T dependence of $1/T_1T$ is attributable to the DOS. The monotonous T dependence at high temperatures is attributed to the DOS involved only at high temperatures. In fact, the photoemission spectroscopy measurements demonstrate that the DOS for LaFeAsO$_{1-x}$F$_x$ decreases with decreasing temperature [23]. This scenario is approved by a quantitative evaluation of $1/T_1T$ just at the plateau. We evaluate $1/T_1T$ using the Korringa relation [24,25] for d-electron alloys [26],

$$1/T_1T = \frac{\pi}{\hbar}(2\hbar\gamma_N A_{\mathrm{hf}})^2 \sum_i n_i(\varepsilon_F)^2 \frac{k_B}{(1-\alpha_Q)^2}, \quad (1)$$

where γ_N, A_{hf}, and $n_i(\varepsilon_F)$ represent the gyromagnetic ratio of ^{75}As (7.292 MHz/10 kOe), the hyperfine coupling constant, and DOS at Fermi surfaces for $i = d_{xy}$, d_{yz}, and d_{zx} orbits, respectively. The factor α_Q is $I\chi(Q)$, where I is the interaction between electrons and $\chi(Q)$ is the susceptibility without the interaction at the dominant wave number Q. The value of A_{hf} has been estimated to be ~ 25 kOe/μ_B from the K-χ plot [27]. The theoretically calculated values of $n_i(\varepsilon_F)$ for $x = 0.20$ at
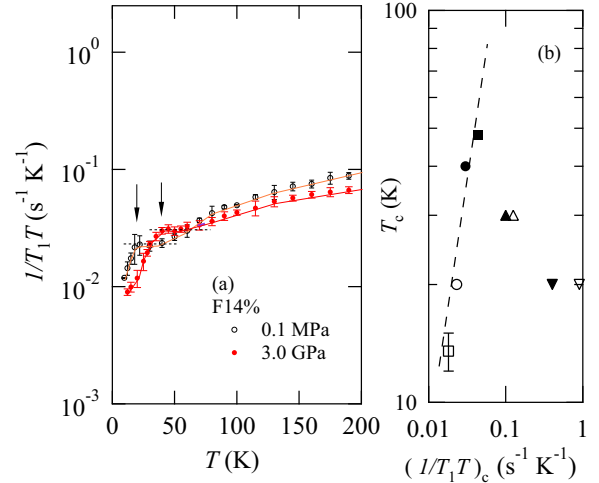

FIG. 5. (a) $1/T_1T$ for LaFeAsO$_{1-x}$F$_x$ ($x = 0.14$). Arrows represent T_c. (b) T_c vs $1/T_1T$ at T_c for LaFeAsO$_{1-x}$F$_x$ and LaFeAsO$_{1-x}$H$_x$. Circles represent the data for $x = 0.14$ (F doping), and squares, regular triangles, and downward pointing triangles represent those for $x = 0.20$, 0.40, and 0.44 (H doping), respectively. Open and solid marks represent the measurements at 0.1 MPa and 3.0 GPa, respectively. The dashed line is a guide to the eye.

ambient pressure are 0.62, 0.92, and 0.92 eV^{-1}, respectively, for $i = d_{xy}$, d_{yz}, and d_{zx} orbits [1]. These values result in $1/T_1T = 4.96 \times 10^{-4} \frac{1}{(1-\alpha_Q)^2}$ (s^{-1} K^{-1}). The plateau of $1/T_1T$ at 0.1 MPa indicates 0.018 s^{-1} K^{-1}, and thus $\alpha_Q = 0.83$. The value is in good agreement with the theoretically estimated value $\alpha_Q = 0.94$ for $x = 0.20$ [28]. At high temperatures, $1/T_1T$ at 3.0 GPa is almost the same as that at 0.1 MPa, which suggests that α_Q is insensitive to pressure and thus α_Q is not a key parameter for increasing T_c.

Figure 5(a) shows the plateaus observed for 14% F-doped samples [13,14]. The T_c enhancement upon applying pressure is highlighted by the dashed arrow in Fig. 4. At 0.1 MPa, the values of $1/T_1T$ at T_c, $(1/T_1T)_c$, are 0.023 and 0.018 s^{-1} K^{-1} for 14% F- and 20% H-doped samples, respectively. At 3.0 GPa, these numbers increase to 0.030 and 0.044, respectively. These data are plotted in Fig. 5(b). The data for 40% and 44% H-doped samples are also plotted for comparison. As seen from the figure, T_c correlates with $(1/T_1T)_c$ only for 14% F- and 20% H-doped samples reflecting $n_i(\varepsilon_F)$ in Eq. (1).

Our results for the La1111 series demonstrate that T_c is not directly affected by AF fluctuations, as clearly seen from Figs. 2(a) and 2(c). Note that the opposite conclusion was derived for the Na111 series, such as NaFe$_{1-x}$Co$_x$As [10]. In the Na111 series, T_c follows AF fluctuations when pressure is applied. At first glance, the results of the Na111 series seem to contradict the results reported herein. One may classify the La1111 series as an exotic and exceptional compound among iron-based pnictides. However, all facts including the La1111 and Na111 series are consistent if the superconductivity is not directly affected by antiferromagnetism or AF fluctuations. For all iron-based pnictides, without exception, $1/T_1T$ becomes stronger as the doping level approaches the AF phase boundary, but the T_c optimal doping level is not always located on the AF phase boundary and depends on the compounds, which causes an apparent discrepancy.

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Roughly speaking, T_c is proportional to the DOS and the pairing interaction. The enhancement of the pairing interaction is hardly expected for the AF-fluctuation-mediated scenario. As another candidate, the orbital-fluctuation-mediated scenario [28] would be promising. In this case, orbital fluctuations are difficult to observe in $1/T_1T$ at a doping level where the structural or nematic phase is absent, and thus the increase in T_c is observable via the DOS alone in $1/T_1T$. To investigate whether the pairing interaction is enhanced simultaneously as well as the DOS, further theoretical investigations are needed, however, the results of Fig. 5(b) may give an important clue.

One may consider another scenario where the pairing interaction originates from AF fluctuations, but AF fluctuations are suppressed at ambient pressure by some competing interactions, such as orbital and/or charge interactions. On the basis of this scenario, T_c could be suppressed and the double-dome structure could be observed, as seen in some high-T_c cuprates. Pressure could nullify the competition, and T_c may increase under pressure even if AF fluctuations are not enhanced. However, the competing orders, which could cause an appreciable suppression of T_c, have not been observed so far in a wide range around $x = 0.14$–0.20. Furthermore, AF fluctuations tend to decrease upon applying pressure, as observed in the poorly F-doped regime [29] or sufficiently H-doped regime [see Fig. 1(c)]. At present, there is no experimental evidence to support this scenario.

In conclusion, we have observed in LaFeAsO$_{1-x}$H$_x$ that T_c for $x = 0.20$ marks a high T_c of 48 K upon applying pressure without the influence of AF fluctuations. For $x = 0.44$ (near the second AF phase boundary), T_c remains unchanged without depending on the magnitude of AF fluctuations. These results suggest that the superconductivity has no direct connection with AF fluctuations. As far as the results of $1/T_1T$ are concerned, the increase in T_c up to 48 K originates from an enhancement of the DOS just above T_c.

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