

Magnetism driven by strong electronic correlations in the heavily carrier-doped iron oxypnictide $\text{LaFeAsO}_{0.49}\text{H}_{0.51}$

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The magnetism of the second antiferromagnetic phase (AF2) arising in the iron-based $\text{LaFeAsO}_{1-x}\text{H}_x$ superconductor for $x \gtrsim 0.4$ was investigated by muon spin rotation measurements under hydrostatic pressure up to 2.6 GPa. The Néel temperature (T_N) obtained for a sample with $x = 0.51$ exhibits considerably greater sensitivity to pressure than that in the pristine antiferromagnetic phase (AF1; $x \lesssim 0.06$). Moreover, while the AF1 phase is always accompanied by the structural transition (from tetragonal to orthorhombic) at a temperature (T_s) which is slightly higher than T_N , the AF2 phase prevails at higher pressures, above ~ 1.5 GPa, where the structural transition is suppressed ($T_s = 0$). These features indicate that the microscopic origin of the AF2 phase is distinct from that of AF1, suggesting that electronic correlation plays an important role in the former phase. We argue that the orbital-selective Mott transition is a plausible scenario to account for the observed pressure dependence of T_N and T_s in the AF2 phase.

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I. INTRODUCTION

Since the discovery of high- T_c superconductivity in iron-based oxypnictides $\text{LnFeAsO}_{1-x}\text{F}_x$ (where Ln denotes a lanthanide) [1–4], the interplay between magnetism and superconductivity in iron-based compounds has been a fascinating topic. While iron is an essential element of the electrically conducting FeAs planes, it usually plays an antagonistic role against superconductivity by bringing about magnetism. As a matter of fact, these compounds under pristine conditions exhibit antiferromagnetic (AF) order below the Néel temperature (T_N), where superconductivity emerges as the AF order is suppressed by carrier doping of the FeAs plane [5].

The emergence of high- T_c superconductivity upon suppression of AF order bears a remarkable similarity to that in cuprates, where the parent compounds are regarded as typical Mott insulators. Although the microscopic mechanism of high T_c is still under debate, it seems now to be commonly presumed that the electronic correlation on the CuO_2 planes (i.e., the strong on-site Coulomb repulsion, which leads to the metal-insulator transition upon half-filling of the $\text{Cu } e_g$ band) is the essential ingredient in cuprates [6]. Meanwhile, the iron-based compounds are distinct from cuprates in that

the pristine compounds exhibit *metallic* AF order (or a spin density wave). Moreover, the AF order is always accompanied by a structural transition at the temperature T_s , which is slightly higher than T_N , suggesting a correlation between the magnetism and the orbital degrees of freedom. These observations lead to the suggestion that spin and/or orbital fluctuations enhanced by the specific Fermi surface topology mediate the Cooper pairing [4].

The recent development of a carrier-doping technique using hydride ion ($\text{LnFeAsO}_{1-x}\text{H}_x$) paved the path to high doping concentrations, up to $x \sim 0.5$, providing the opportunity to investigate the relationship between magnetism and superconductivity over an unprecedented range of x [7–10]. As shown in Fig. 1, the extended doping in the case of $\text{Ln} = \text{La}$ [9] led to the discovery of a new superconducting phase (SC2) marked by a second peak of T_c around $x \sim 0.36$ (with a dome-like x dependence of T_c) and an associated AF phase (AF2) that emerges at $x \gtrsim 0.4$ in place of the SC2 phase, establishing a novel bipartite phase diagram together with the pristine AF phase ($x \lesssim 0.05$; denoted AF1) and the known superconducting phase (SC1; accompanying another T_c dome with a peak around $x \sim 0.1$), which is separated by a valley of T_c near $x \sim 0.2$ [11–13].

As inferred from previous studies on a sample with $x = 0.51$, the AF2 phase is characterized by a unique structural modulation and magnetic structure different from that of the AF1 phase [12]. Although the existence of the AF2 phase adjacent to SC2 in the bipartite phase diagram suggests a causal

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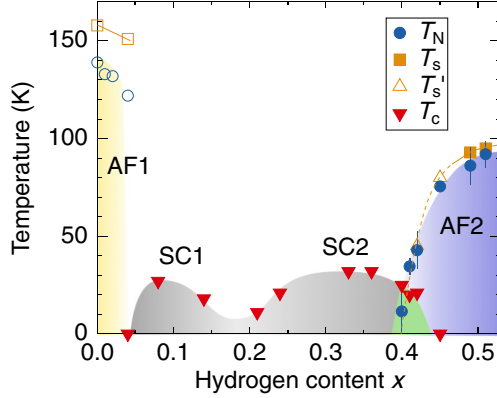


FIG. 1. Electronic phase diagram of $\text{LaFeAsO}_{1-x}\text{H}_x$ along H content x , where AF1/2 and SC1/2 denote antiferromagnetic and superconducting phases [12]. The experimentally determined Néel temperature T_N , structural transition temperature T_s , and superconducting transition temperature T_c are represented by circles, triangles, and inverted triangles, respectively.

relationship between these two phases, the subtle difference between the AF1 and the AF2 phases hints at a possible distinction in the mechanism of superconductivity between the corresponding SC phases.

According to earlier resistivity measurements, the T_c domes of the SC1 and SC2 phases tend to overlap when a hydrostatic pressure is applied, merging into a single dome at 6 GPa, with a maximum T_c of 52 K [14]. Furthermore, it is inferred from recent synchrotron x-ray diffraction measurements that the structural transition ($T_s = 95$ K for $x = 0.51$) under ambient pressure is suppressed ($T_s = 0$) under the relatively low pressure of 1.5 GPa [15]. These features suggest a strong connection between the T_c and the lattice structure, providing an important clue for understanding the mechanism behind high- T_c superconductivity in $\text{LaFeAsO}_{1-x}\text{H}_x$. This connection naturally raises questions regarding the interrelationship between lattice structure and magnetism in the AF2 phase. To address this issue, we conducted muon spin rotation (μSR) measurements under hydrostatic pressures on a $\text{LaFeAsO}_{1-x}\text{H}_x$ sample with $x = 0.51$ situated in the AF2 phase.

II. EXPERIMENTAL METHODS

A polycrystalline sample identical to that used for the x-ray diffraction experiment ($x = 0.51$) [15] was adopted for the μSR experiment to avoid ambiguity due to possible fluctuation of the doping concentration; the details of the sample preparation are reported in Ref. [7]. Conventional μSR measurements under hydrostatic pressure were performed using the general-purpose decay-channel spectrometer of the Swiss Muon Source facility at the Paul Scherrer Institute, Switzerland. A powder sample (~ 1.5 g) was pressurized within a cylindrical space with a diameter of 5.9 mm using Daphne oil 7373 as pressure-transmitting medium. The sample was sealed by a double-wall pressure cell made of NiCrAl and MP35N alloys [16]. The exact pressure inside the cell was determined from the superconducting transition temperature T_c of a small piece of indium, which was also mounted on

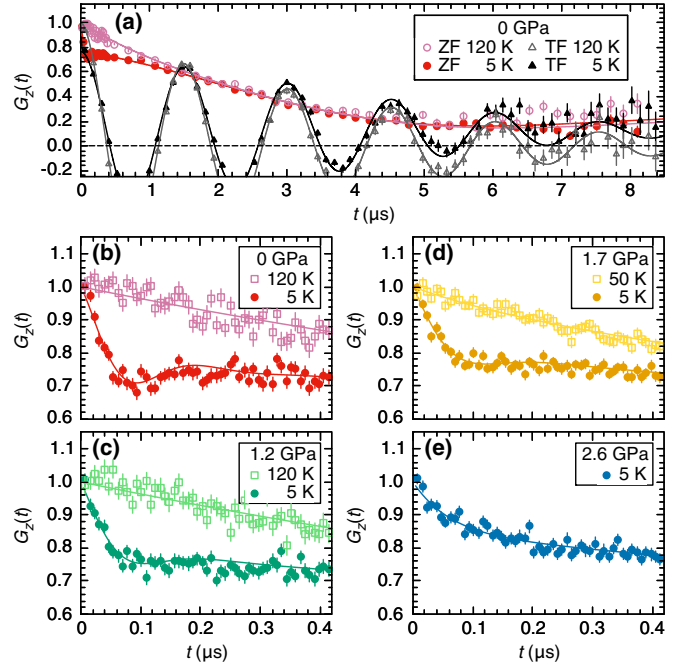


FIG. 2. (a) Typical ZF- and TF- μSR spectra of $\text{LaFeAsO}_{1-x}\text{H}_x$ with $x = 0.51$ (normalized to the value at $t = 0$) measured at temperatures above (120 K) and below (5 K) T_N under ambient pressure. Spectra for TF = 5 mT are partially represented for clarity. (b-e) ZF- μSR spectra measured under (b) ambient pressure, (c) 1.2 GPa, (d) 1.7 GPa, and (e) 2.6 GPa. Open and filled symbols represent the spectra above T_N and those at the lowest temperature, respectively. Solid curves show the best fits using Eq. (1).

the same sample mount space [17]. A muon beam with a momentum of 99.25 MeV/c was irradiated to penetrate the thick wall of the pressure cell and to maximize the number of muons stopped in the sample space. The pressure cell was loaded onto a cryostat under He gas flow to monitor the time-dependent μSR spectra [positron decay asymmetry $A_z(t)$] under a zero field (ZF) or a transverse external field (TF; 5 mT) in the 5–140 K temperature range.

III. RESULTS

In the high-pressure setup, the μSR spectra consist of two signal components, one corresponding to muons stopped in the sample and the other to muons from the pressure cell. In ZF, these are described by

$$A_0 G_z(t) = A_{\text{smp}} G_{\text{smp}}^z(t) + A_{\text{pc}} G_{\text{pc}}^z(t),$$

$$G_{\text{pc}}^z(t) = G_{\text{KT}}^z(\Delta_{\text{ZF}}, t) \exp(-\lambda_{\text{pc}} t), \quad (1)$$

where $G_{\text{smp}}^z(t)$ and $G_{\text{pc}}^z(t)$ represent the time evolution of the muon spin polarization in the sample and in the pressure cell, respectively, with their respective partial asymmetry being A_{smp} and A_{pc} ($A_0 = A_{\text{smp}} + A_{\text{pc}}$). $G_{\text{pc}}^z(t)$ is known to be described by the static Kubo-Toyabe relaxation function $G_{\text{KT}}^z(\Delta_{\text{ZF}}, t)$ multiplied by an exponential damping at rate λ_{pc} to empirically account for the depolarization in the cell, with the term Δ_{ZF} representing the line width caused by nuclear magnetic moments [16].

Figure 2(a) shows typical examples of ZF- and TF- μSR spectra under ambient pressure. The ZF spectrum at 5 K

($<T_N$) is dominated by a slow Gaussian-like depolarization for $t \geq 0.2 \mu\text{s}$, indicating that the line shape at later times is predominantly determined by $G_{\text{KT}}^z(\Delta_{\text{ZF}}, t)$ for muons stopped in the nonmagnetic pressure cell. Meanwhile, the slow exponential-like depolarization at 120 K ($>T_N$) represents the behavior of $G_{\text{smp}}^z(t)$ overlapped with $G_{\text{KT}}^z(\Delta_{\text{ZF}}, t)$, which originates from residual iron impurities known to exist in the present specimen [18]. Considering that the situation resembles a canonical dilute spin glass (e.g., AuFe) [19], we attribute this behavior to a spin-glass-like impurity phase that coexists with the AF phase below T_N .

It is clear in the magnified Figs. 2(b)–2(e) that the spectra at 5 K exhibits fast damping precession under ambient pressure and that the precession becomes obscure with increasing pressure, suggesting a decrease in the internal field (and/or the fractional yield) of the AF phase probed by the muon because of progressive suppression of the magnetic correlation. Considering these observations, the spectra were analyzed by χ^2 -minimization curve fitting using Eq. (1) and the following expression for $G_{\text{smp}}^z(t)$:

$$G_{\text{smp}}^z(t) = wG_{\text{mag}}(t) + (1 - w)G_{\text{sg}}(t).$$

Here, $G_{\text{mag}}(t)$ represents a component exhibiting relatively well-defined AF order with the volume fraction w , and $G_{\text{sg}}(t)$ accounts for the remaining nonmagnetic ($T_N \sim 0$) fraction dominated by spin-glass-like behavior. Assuming quasistatic magnetism, $G_{\text{mag}}(t)$ and $G_{\text{sg}}(t)$ are approximated by

$$G_{\text{mag}}(t) = \frac{1}{3} + \frac{2}{3} \cos(2\pi ft + \phi)e^{-\lambda t}, \quad (2)$$

$$G_{\text{sg}}(t) = \frac{1}{3} + \frac{2}{3}(1 - \lambda t)e^{-\lambda t}, \quad (3)$$

where the first term represents the fraction of muons subject to the component of the quasistatic local field B_{loc} parallel to the initial muon spin direction $\hat{z}$ and is 1/3 for powder samples, and the second term represents that for $B_{\text{loc}} \perp \hat{z}$. In $G_{\text{mag}}(t)$, the muon spin exhibits precession with a frequency $f = \gamma_\mu B_{\text{loc}}/2\pi$ (with $\gamma_\mu = 135.538 \times 2\pi \text{ MHz/T}$ being the muon gyromagnetic ratio) for nonzero B_{loc} . We adopted the Lorentzian Kubo-Toyabe function for $G_{\text{sg}}(t)$ to describe the spin-glass-like behavior [19], where λ was $\sim 0.5 \mu\text{s}^{-1}$ for the relevant temperature range. In the quasistatic magnetic phase, the local field probed by muons is mainly determined by a vector sum of the magnetic dipolar field of the Fe atoms,

$$B_{\text{loc}} = \left| \sum_i \hat{A}_i \mu_i \right|, \quad (4)$$

where μ_i is the magnetic moment of the i th Fe located at distance $\mathbf{r}_i = (x_i, y_i, z_i)$ from the muon site, and

$$\hat{A}_i = A_i^{\alpha\beta} = \frac{1}{r_i^3} \left(\frac{3\alpha_i\beta_i}{r_i^2} - \delta_{\alpha\beta} \right) \quad (\alpha, \beta = x, y, z)$$

is the dipolar tensor. Although B_{loc} is a scalar quantity, it provides a strong criterion to verify the consistency among the muon site(s), magnetic structure, and Fe-moment magnitude inferred from other experimental techniques (see below). In the following curve-fit analysis, the fraction of muons stopped in the sample, $f_s \equiv A_{\text{smp}}/(A_{\text{smp}} + A_{\text{pc}})$, and the total asymmetry A_0 at the four applied pressures are fitted simultaneously in order to impose a common value for all temperatures,

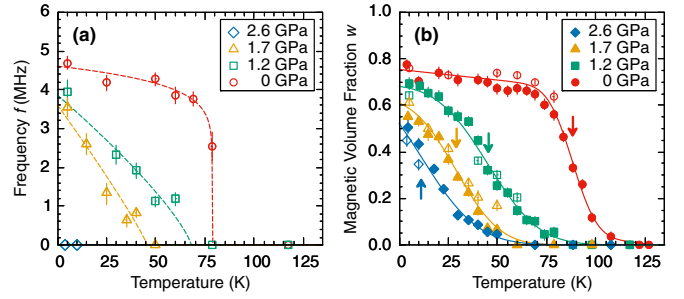


FIG. 3. Temperature dependence of (a) f and (b) w under different pressures. Filled (open) symbols indicate values obtained from TF- μ SR (ZF- μ SR) measurements, respectively. Dashed lines in (a) are a guide for the eye. Solid curves in (b) are the best-fit curves used to extract the magnetic transition temperature T_N , denoted by filled arrows, as described in the text.

yielding $f_s = 0.45$ – 0.52 and $A_0 \simeq 0.28$. For the signal coming from the pressure cell, Δ_{ZF} was fixed to the value obtained by interpolation at each temperature, and λ_{pc} was fixed to $0.04 \mu\text{s}^{-1}$ since it is known to be almost unchanged down to 1 K [16], which is far below the lowest temperature attained in our study.

The temperature dependence of the frequency f under four different pressures is shown in Fig. 3(a). We note that f was fixed to 0 in analyzing the spectrum under 2.6 GPa because no clear precession signal was discernible [see Fig. 2(e)]. The decrease in f at the lowest temperature with increasing applied pressure indicates that B_{loc} is reduced accordingly. Assuming that the magnetic structure is unchanged, this suggests that the magnitude of the Fe moments [which is proportional to B_{loc} ; see Eq. (4)] decreases for increasing pressure. (For a discussion of the possible broadening of B_{loc} induced by pressure, see Sec. IV B.)

The fact that a long-lived precession with a frequency proportional to the external field B_0 is observed above T_N for the entire asymmetry of TF- μ SR spectra [including that corresponding to A_{smp} ; see Fig. 2(a)] indicates that the mean field in the spin-glass-like phase is much weaker than B_0 , while those in the magnetic phase are depolarized rapidly because of the distribution of B_{loc} originating from the magnetic order,

$$B_{\text{loc}} = \left| \sum_i \hat{A}_i \mu_i + B_0 \right|.$$

The upward shift of the 5 K spectrum shown in Fig. 2(a) derives from the first term in Eq. (2), which reflects the muons subject to the total local field parallel to $\hat{z}$. The μ SR spectra under a TF of 5 mT were analyzed using Eq. (1) by replacing $G_{\text{pc}}^z(t)$ and $G_{\text{sg}}^z(t)$, respectively, with

$$G_{\text{pc}}^z(t) = \exp(-\lambda_{\text{pc}}t) \exp(-\sigma_{\text{TF}}^2 t^2/2) \cos(\gamma_\mu B_0 t + \phi),$$

$$G_{\text{sg}}^z(t) = \exp(-\lambda t) \cos(\gamma_\mu B_0 t + \phi),$$

where σ_{TF} is the relaxation rate in TF- μ SR measurements caused by the nuclear magnetic moments in the pressure cell and ϕ is the initial phase of the precession. σ_{TF} and λ_{pc} were also fixed to the values reported in Ref. [16], as described above. The temperature dependence of w under different pressures is shown in Fig. 3(b), where the onset

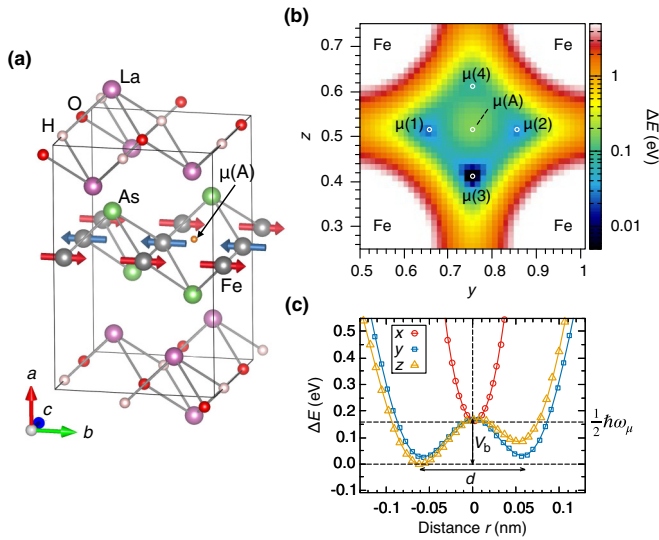


FIG. 4. (a) Crystal and magnetic structure of LaFeAsO_{0.49}H_{0.51}. The possible muon site $\mu(A)$ is represented by the small orange circle. Red and blue arrows at Fe atoms represent magnetic moments along the y axis. (b) Contour plot of ΔE in the yz plane for $x = 0.56$ in the Aem2 orthorhombic phase of LaFeAsO_{1-x}H_x for $x = 0.51$. Regions of $\Delta E > 5$ eV are shown in white for clarity. $\mu(1)$ – $\mu(4)$ and $\mu(A)$ denote four ΔE minima and the central position (0.56, 0.75, 0.51). (c) ΔE profiles along the x, y, and z axes passing through the $\mu(A)$ site as functions of the distance r from the $\mu(A)$ site.

temperature decreases with increasing pressure. At the lowest temperature, w decreases with increasing pressure, indicating that the volume fraction of the AF2 phase decreases. These results were used to determine the mean value and uncertainty of T_N (see below).

IV. DISCUSSION

A. Muon site

In evaluating the magnetism of the AF2 phase based on the μ SR results, it is important to have a good estimate of the muon site. Since muons behave as a pseudohydrogen in matter, the variation in total energy upon the inclusion of H estimated by density functional theory (DFT) calculations serves as a guide to narrow down the candidate muon sites. The most probable site is inferred from the consistency of the B_{loc} calculated using Eq. (4) for the candidate sites with that measured. We calculated the total energy for interstitial H in the Aem2 orthorhombic phase using the OPENMX code, which is based on the generalized gradient approximation to DFT and the norm-conserving pseudopotential method [20]. We used a cutoff energy of 150 Ry and a $3 \times 3 \times 3$ mesh at the K point with the experimentally obtained lattice constant [12].

Figure 4(b) shows the variation of the total energy $\Delta E = E_{\text{tot}}(\mathbf{r}) - E_{\text{min}}$ versus the H position $\mathbf{r}$, where E_{min} is the global minimum of $E_{\text{tot}}(\mathbf{r})$. As shown by $\mu(1)$ – $\mu(4)$, four minima of ΔE are revealed around the central position $\mu(A)$, which corresponds to the saddle point. The slightly different values of ΔE at the minima may be attributed to the off-center deformation of the FeAs₄ tetrahedron in the Aem2 structure. The

TABLE I. Simulated internal magnetic field $B_{\text{loc}}^{\text{sim}}$ at each muon site induced by the magnetic moments of the Fe atoms. The value of $B_{\text{loc}}^{\text{sim}}$ for $x = 0.51$ at $\mu(A)$ represents the range of $B_{\text{loc}}^{\text{sim}}$ around the $\mu(A)$ position within $\Delta r = 5.8$ pm on the bc plane [see Fig. 5(b)].

Muon site	Magnetic field (mT)	
	$x = 0$ ($ m = 0.63 \mu_B$ [21])	$x = 0.51$ ($ m = 1.21 \mu_B$ [12])
$\mu(A)$	144.3	0–49.7
$\mu(1), \mu(2)$	259.2	467.0
$\mu(3)$	306.6	371.9
$\mu(4)$	306.6	319.9
Experiment	173.2 [28,29]	37.7 ± 0.5 [12]

distance from $\mu(A)$ is 0.056, 0.059, and 0.053 nm for $\mu(1, 2)$, $\mu(3)$, and $\mu(4)$, respectively. Figure 4(c) represents the ΔE profiles along the x, y, and z axes passing through the $\mu(A)$ site. ΔE rapidly increases along the x axis (perpendicular to the FeAs plane) when the distance $|r|$ from $\mu(A)$ is increased, indicating that muons may be confined within the yz plane. However, along the y and z axes, ΔE exhibits an asymmetric double-well potential structure with the maximum potential barrier $V_b = 169.1$ meV located between $\mu(3)$ and $\mu(4)$ at a distance $d = 0.112$ nm. In the harmonic approximation of the sinusoidal potential curve, the energy level splitting $\hbar\omega_\mu$ for the bound-state muon is derived from the relation $\omega_\mu^2 = 2\pi^2 V_b / m_\mu d^2$, where $m_\mu = 105.658$ MeV/ c^2 is the muon mass. The corresponding zero-point energy $\frac{1}{2}\hbar\omega_\mu$ in this approximation is estimated to be 156.5 meV, comparable to V_b . This suggests that the muon is virtually located at the $\mu(A)$ site for a time longer than ω_μ^{-1} , constituting a typical example of the isotope effect between muon and hydrogen.

The muon site was identified by comparing the B_{loc} obtained from the ZF- μ SR measurements in the magnetically ordered phase with that calculated using Eq. (4) for the candidate sites, summing the Fe moments located within 10 nm from the muon site. The $B_{\text{loc}}^{\text{sim}}$ values calculated at each muon site for $\mu(A)$ and the $\mu(1)$ – $\mu(4)$ minima, using the reported crystal and magnetic structure for $x = 0$ [21] and $x = 0.51$ [12], are summarized in Table I. For $x = 0$, although $B_{\text{loc}}^{\text{sim}}$ agrees with the value obtained experimentally at $\mu(A)$, it is larger than B_{loc} by a factor of 1.5–1.8 at the $\mu(1)$ – $\mu(4)$ sites. Similarly, for $x = 0.51$, $B_{\text{loc}}^{\text{sim}}$ at $\mu(1)$ – $\mu(4)$ is much larger than B_{loc} , by a factor of ~ 10 . The simulated value at $\mu(A)$ for $x = 0.51$ represents the range of $B_{\text{loc}}^{\text{sim}}$ within $\Delta r = 5.8$ pm from the $\mu(A)$ position, where Δr corresponds to the resultant mesh size for real space in our DFT calculation. Although $B_{\text{loc}}^{\text{sim}}$ changes steeply, from 0 to 49.7 mT, even for such a small mesh size [see Fig. 5(b)], its simple average within $\Delta r \simeq 8$ pm yields ~ 38 mT, in close agreement with the experimental value. These results indicate that the muon occupies $\mu(A)$ for $x = 0$ and 0.51 because of its small mass, unlike hydrogen for $x = 0.1$ [22].

B. Magnitude and distribution of B_{loc} below T_N

Figures 5(a) and 5(b) represent the simulated internal magnetic field distribution around muon site $\mu(A)$ on the

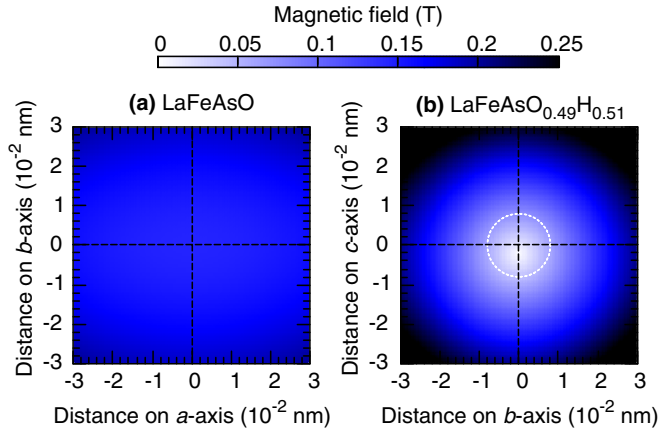


FIG. 5. (a) In-plane $B_{\text{loc}}^{\text{sim}}$ magnetic field distribution around the muon site within 0.03×0.03 nm at (a) $z = 0.573$ for LaFeAsO and (b) $x = 0.56$ for LaFeAsO_{0.49}H_{0.51}. Regions where the magnetic field is >0.25 T are shown in black for clarity. The dotted white circle in (b) represents the region within $\Delta r \simeq 8$ pm where the field is averaged to yield $B_{\text{loc}}^{\text{sim}} \simeq 38$ mT.

plane parallel to the FeAs layer for $x = 0$ and $x = 0.51$, respectively. Although the profile of the field around muon site $\mu(A)$ (at the center of the graph) is nearly independent of position for $x = 0$, a steep profile is revealed for $x = 0.51$. This indicates that a tiny displacement of the muon site does not affect the field profile probed by muons for $x = 0$, whereas a strong dependence is expected for $x = 0.51$. This is especially important when considering that a muon site displacement is more probable for $x = 0.51$ than for $x = 0$ because the substituted hydrogen randomly occupies the oxygen site. Thus, at $x = 0.51$, muons may probe the broad field profile, causing the fast depolarization spectrum below T_N shown in Fig. 2(b). The underestimated value of $B_{\text{loc}}^{\text{sim}}$ at $\mu(A)$ for $x = 0.51$ may be attributed to this broad profile.

We examined the influence of pressure on the local field by calculating $B_{\text{loc}}^{\text{sim}}$ for the lattice constants reduced by external pressure reported in Ref. [15]. According to a recent NMR experiment, the AF2 phase is stable below 2 GPa because of a large gap [23], suggesting that the magnetic structure is unchanged. Under the further assumption that the muon site is also intact with pressure, $B_{\text{loc}}^{\text{sim}}$ at 2 GPa increases by $\sim 2\%$, as expected from the definition of $B_{\text{loc}}^{\text{sim}}$ provided by Eq. (4). This trend is opposite to the experimental results, as shown in Fig. 3(a), necessitating other causes for the observed decrease in B_{loc} with increasing pressure. In this regard, it is interesting to note that a reduction of the Fe-moment magnitude under pressure is predicted by theoretical studies of LaFeAsO [24–26], which is understood as a result of the increased energy band width. Our estimation indicates that the reduction of the Fe moment by $\sim 25\%$ is sufficient to account for the experimental result around 2 GPa.

We also draw attention to the fact that the μSR spectrum for $x = 0.51$ at the lowest temperature and under ambient pressure is dominated by one precession term with fast depolarization. This is in sharp contrast to the $x = 0$ case, where the spectrum at the lowest temperature is well reproduced by the sum of the higher-frequency (lower-frequency)

term of $f \sim 23$ MHz ($f \sim 3$ MHz) with the fractional ratio of 7:3 [27–30]. This indicates the presence of two magnetically inequivalent muon sites. The second-lowest-energy site, $\mu(B) = (0.09, 0.37, 0.11)$ for $x = 0.51$, is assumed to be in the La-O/H layer with distance $r_{\text{O}\mu} \simeq 0.13$ nm from the nearest oxygen. This suggests the formation of a local state bound to oxygen, as that empirically established in many oxides (typical $r_{\text{O}\mu} \simeq 0.1$ nm).

We calculated the local field at the $\mu(B)$ site, finding $B_{\text{loc}}^{\text{sim}} = 0.8$ mT. The smaller B_{loc} is ascribed to the greater $|r_i|$ for the $\mu(B)$ site from Fe moments [see Eq. (4)], where the distances from the nearest Fe atom to the $\mu(A)$ and $\mu(B)$ sites are 0.20 and 0.37 nm, respectively. Although the corresponding signal at $f = 0.1$ MHz has a magnitude that can be detected by conventional μSR measurements, under our experimental conditions ($f_s \sim 0.5$) the low fractional ratio (0.3–0.4) for the $\mu(B)$ site [27,28,30] and the signal-to-noise ratio of ~ 1 impede the separation of this component from the slowly depolarizing ones, i.e., $G_{\text{sg}}(t)$ and/or $G_{\text{pc}}^z(t)$ in Eq. (1). This may result in underestimation of w in Fig. 3(b). Actually, the nonmagnetic volume fraction deduced under ambient pressure at $T \rightarrow 0 = 1 - w(T = 0) \simeq 0.25$ could correspond to the fraction $f_s(B)$ of muons stopped at $\mu(B)$. Supporting this, we note that its value is comparable to the expected value of $f_s(B)$ in the present setup, $f_s(B) = (0.3\text{--}0.4) \times f_s = 0.13\text{--}0.18$ with $f_s = 0.45$. We also refer to the possibility that B_{loc} exhibits a broader distribution under pressure, which will result in the absence of any detectable oscillation for the spectrum stemming from the $\mu(B)$ site.

C. Magnetism vs lattice structure

The solid curves in Fig. 3(b) are the best fits using the equation

$$w(T) = \frac{1}{2}w(0) \left[1 - \text{erf} \left(\frac{T - T_N}{\sqrt{2}\Delta T_N} \right) \right],$$

in which a Gaussian distribution of width ΔT_N is assumed around the average transition temperature T_N (a linear term was added only for ambient pressure data to account for the gradual increase with decreasing temperature) [31]. The obtained T_N and that for $x = 0$ [30] are shown in Fig. 6 as a function of the pressure, along with the structural transition temperature T_s determined for the same sample in Ref. [15]. The width ΔT_N is represented as error bars for T_N , although T_N itself is well determined within an error of ~ 1 K, except at the highest pressure. The large error bars resulting for $T_N(2.6 \text{ GPa}) \sim 6$ K may originate from the strong temperature-dependent behavior of w in the lowest-temperature region shown in Fig. 3(b).

It is remarkable that the structural transition to the orthorhombic phase with decreasing temperature is suppressed near 1.5 GPa ($T_s \rightarrow 0$), whereas the AF2 phase survives even under 2.6 GPa. This is in sharp contrast with the AF1 phase, in which T_N is always below T_s , indicating that the magnetic order of the AF2 is induced by a purely electronic mechanism. According to a theoretical study based on molecular orbitals [32], the electronic state of the AF2 phase is understood through an orbital-selective Mott transition, where Fe-3d_{xy}

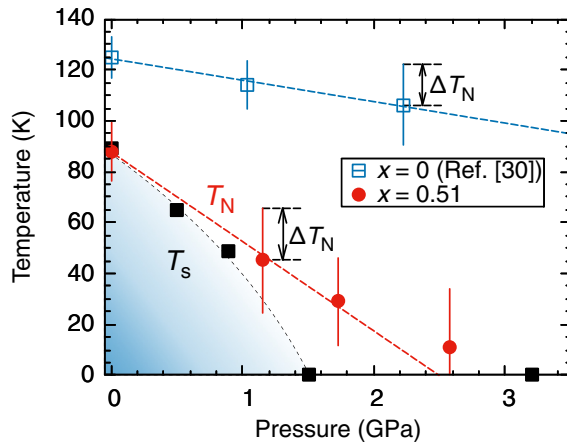


FIG. 6. T_N and ΔT_N , as a function of the external pressure, and T_s data from Ref. [15]. The transition width ΔT_N is represented as error bars for T_N . Data represented by blue squares are cited from Ref. [30]. Dashed lines and the blue region are guides for the eye.

becomes half-filled when x is increased. This situation is similar to that of the e_g orbital in pristine cuprate compounds, implying that the AF order of the AF2 phase is induced by electronic correlation. The fact that T_N is independent of T_s in the AF2 phase supports the above expectation. The pressure dependence of T_N is also understood within this scenario (see below).

Here, it may be worth mentioning that the appearance of the AF order that precedes the structural transition with decreasing temperature bears a remarkable similarity to the so-called electronic nematicity revealed in BaFe_2As_2 (122) family compounds, where the isovalent substitution of As with P induces a unidirectional self-organized state that breaks the rotational symmetry of the underlying lattice above T_s [33]. In addition, the coexistence of the AF2 phase and SC2 phase observed over the finite doping range of $0.4 \leq x \leq 0.45$ [12] comprises yet another parallelism with the 122 family [34], hinting at the importance of electronic correlation in the latter compounds.

The sensitivity of magnetism to pressure in the AF2 phase is quantitatively described by the gradient $dT_N/dp = -35.5 \pm 0.6 \text{ K GPa}^{-1}$, which is much greater than the $-8.5 \pm 0.1 \text{ K GPa}^{-1}$ of the AF1 phase ($0 \leq x \leq 0.06$) [30], indicating that the AF2 phase is more susceptible to pressure than AF1. This contrast may originate from the different mechanism of magnetic order. In the AF1 phase, the AF order is induced by nesting of the Fermi surface, as is concluded in a theoretical study on LaFeAsO ($x = 0$; AF1 phase) reporting that the nesting condition is almost unchanged between 0.1419 and 0.120 nm³ (corresponding to -7 and 10 GPa), indicating the robustness of the AF1 phase against pressure

[24]. On the other hand, monotonic degradation of the nesting with doping due to the expansion of the electron Fermi surface at the M point [35] disfavors a similar scenario for the AF2 phase.

It is reported that the energy gap between bonding and antibonding orbitals upon Fe $3d$ -As $4p$ hybridization in the low- x region decreases when the height of the As ion from the Fe plane h_{As} increases. As h_{As} increases (which is equivalent to an increase in x), the nonbonding Fe- $3d_{xy}$ orbital becomes half-filled, resulting in the orbital-selective Mott state. Because external pressure induces a considerable decrease in h_{As} in $\text{LaFeAsO}_{1-x}\text{H}_x$ [15], the fragility of the AF2 phase against pressure is readily understood within the scenario of an orbital-selective Mott transition [32]. In fact, for $x = 0.51$ and at ambient pressure, $h_{\text{As}} = 0.1413 \text{ nm}$, decreasing to 0.1375 nm under 2.2 GPa. This value is comparable to that for $x \sim 0.4$, where the AF2 phase is nearly suppressed [12,15].

Finally, we note that the strong pressure dependence of T_N combined with the spatial inhomogeneity of pressure in the sample space originating from a partial nonhydrostaticity may also contribute to the nonmagnetic phase ($1 - w$) below T_N , the confirmation of which remains as a future task using a more refined μSR sample environment for high pressure.

V. SUMMARY AND CONCLUSION

To summarize, our μSR study of $\text{LaFeAsO}_{1-x}\text{H}_x$ with $x = 0.51$ under external pressure revealed that the AF2 phase survives under a pressure as high as 2.6 GPa, far beyond the pressure where the structural transition to the orthorhombic phase is suppressed. The AF2 phase with $x = 0.51$ is more susceptible to pressure than the AF1 phase with $x = 0$, suggesting a different magnetic ordering mechanism. Considering theoretical works, the AF1 phase is robust against external pressure because the nesting of the Fermi surface that induces it is nearly independent of the pressure. In contrast, the AF2 phase is understood through the orbital-selective Mott state, in which the height parameter h_{As} plays an essential role. Because h_{As} decreases when external pressure is applied, the AF2 phase is sensitive to the latter.

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