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Nuclear magnetic resonance on $LaFeAsO_{0.4}H_{0.6}$ at 3.7 GPa

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A prototypical electron-doped iron-based superconductor $LaFeAsO_{1-x}H_x$ undergoes an antiferromagnetic (AF) phase for $x \geq 0.49$. We have performed nuclear magnetic resonance (NMR) measurements on $LaFeAsO_{0.4}H_{0.6}$ at 3.7 GPa to investigate the magnetic properties in the vicinity of a pressure-induced quantum critical point (QCP). The linewidth of ^{1}H-NMR spectra broadens at low temperatures below 30 K, suggesting that the spin moments remain ordered at 3.7 GPa. The coexistence of gapped and gapless spin excitations was confirmed in the ordered state from the relaxation time T_1 of ^{75}As. The pressure-induced QCP is estimated to be 4.1 GPa from the pressure dependence of the gapped excitation.

I. Introduction

A prototypical electron-doped iron-based pnictide $LaFeAsO_{1-x}H_x$ ($0 \leq x \leq 0.6$) exhibits unique electronic properties in a heavily carrier-doped regime: a superconducting (SC) phase with double-domes structure expands in a wide regime ($0.05 < x < 0.49$) [1] and an antiferromagnetic (AF) phase manifests itself by further H doping ($0.49 \leq x$) [2–4]. Band calculations show that both Fermi surfaces and nesting vectors change by H doping: the two hole pockets present at Γ point in the lightly H-doped regime almost disappear in the heavily H-doped regime [5,6]. The change in the nesting vectors due to H doping would cause a change in wave-vector (q) dependent spin susceptibility $\chi(q,\omega)$ and would allow for the appearance of two AF phases in the lightly and heavily H-doped regimes.

The AF phase in the heavily H-doped regime is strongly suppressed upon applying pressure [7]. We have performed nuclear magnetic resonance (NMR) measurements on $LaFeAsO_{0.4}H_{0.6}$ at 3.7 GPa, and we have found that the spin excitation gap appearing at the AF phase vanishes at around 4.1 GPa. We have investigated the magnetic properties in the vicinity of a pressure-induced quantum critical point (QCP)($\simeq$4.1 GPa).

II. Experimental apparatuses and conditions

A pressure of 3.7 GPa was applied using a NiCrAl-hybrid clamp-type pressure cell as shown in Fig. 1 [8]. We have used a mixture of Fluorinert FC-70 and FC-77 as the pressure-transmitting medium. A coil wounded around the powder samples and an optical fiber with the Ruby powders glued on top

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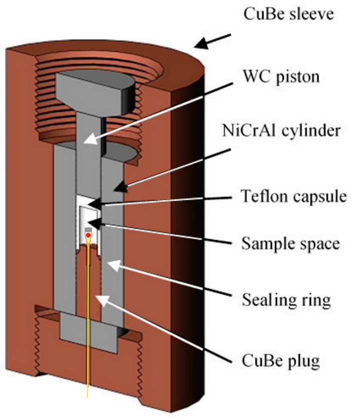

Figure 1: A NiCrAl-hybrid clamp-type pressure cell [8]. A coil wounded around the powder samples and an optical fiber with the Ruby powders were inserted into the sample space.

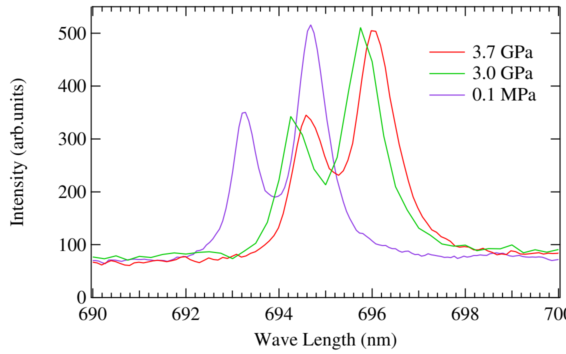

Figure 2: Ruby fluorescence spectra. The smaller and larger peaks correspond to the R2 and R1 transitions, respectively.

were inserted into the sample space of the pressure cell [8]. The size of the coil was 2.4 mm in diameter and 3.5 mm in length, and the number of windings was 18 turns. The pressure was monitored through Ruby fluorescence measurements. The R1 and R2 lines at ambient pressure, 3.0 and 3.7 GPa are shown in Fig. 2. The wavelength of the R1 or R2 peak shifts linearly with respect to pressure. The shift of the wavelength $\Delta\lambda$ satisfies the relation $P(\mathrm{GPa})=\Delta\lambda(nm)/0.365$.

NMR measurements for the powder samples were acquired using a conventional coherent-pulsed NMR spectrometer. The relaxation rate $(1/T_1)$ was measured using a conventional saturation-recovery method for the samples whose FeAs planes are parallel to the applied field.

III. Experimental results

i. ^{1}H-NMR spectra

^{75}As($I = 3/2$)-NMR spectra broaden due to the nuclear quadrupole interaction, which makes difficult to investigate the antiferromagnetic (AF) state. However, ^{1}H($I = 1/2$) is free from the nuclear quadrupole interaction. Therefore, the ^{1}H signal is narrow at a paramagnetic state, and the broadening in the AF phase directly reflects the mag-

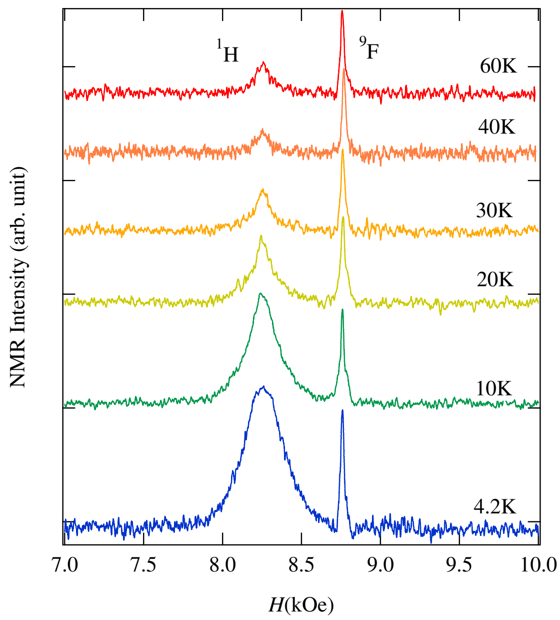

Figure 3: ^{1}H-NMR spectra for $LaFeAsO_{0.4}H_{0.6}$ measured at 3.7 GPa and 35.1 MHz. The ^{9}F signal originates from the pressure-transmitting medium, a mixture of Fluorinert FC-70 and FC-77.

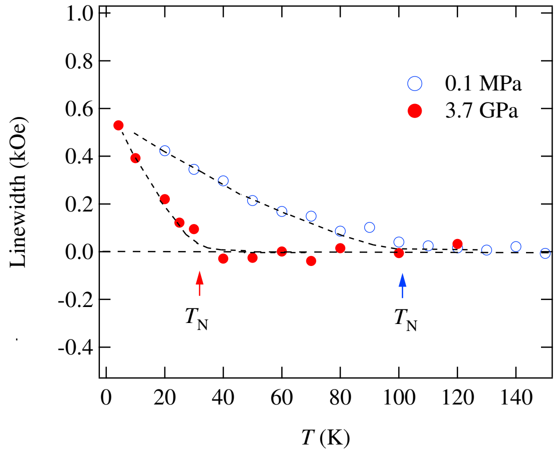

Figure 4: The increase in ^{1}H linewidth due to the ordered spin moments. T_N represents the antiferromagnetic (AF) transition temperature.

nitude of the spin moments. Figure 3 shows ^{1}H-NMR spectra measured at 3.7 GPa and 35.1 MHz. The sharp signal of ^{9}F originates from the pressure-transmitting medium mentioned above. The temperature dependence of the linewidth is shown in Fig. 4 together with the data at ambient pressure [2, 4]. The onset of the broadening in Fig. 4 corresponds to the AF transition temperature (T_N). The maximum spin moment is estimated to be 1.80 μ_B [4]. As seen in Fig. 4, T_N is about 100 K at ambient pressure and decreases to 30 K at 3.7 GPa. The pressure-induced QCP is expected at a much higher pressure regime.

ii. $1/T_1T$ for ^{75}As

The relaxation rate divided by temperature $1/T_1T$ provides a measure of low-energy spin fluctuations. In general, neglecting the wave-number (q) dependence of the hyperfine coupling constant, $1/T_1T$ is proportional to the imaginary part of the susceptibility: $1/T_1T \propto \Sigma_q Im\chi(q,\omega)/\omega$ where ω represents a NMR frequency. ^{75}As is preferred to ^{1}H for T_1 measurements, because FeAs layers are hardly affected by the random distrubution of hydrogen in $LaO_{1-x}H_x$ layers. Furthermore, owing to the nuclear quadrupole interaction, one can pick up the ^{75}As signals coming from the powders whose FeAs planes are parallel to the applied field. Figure 5

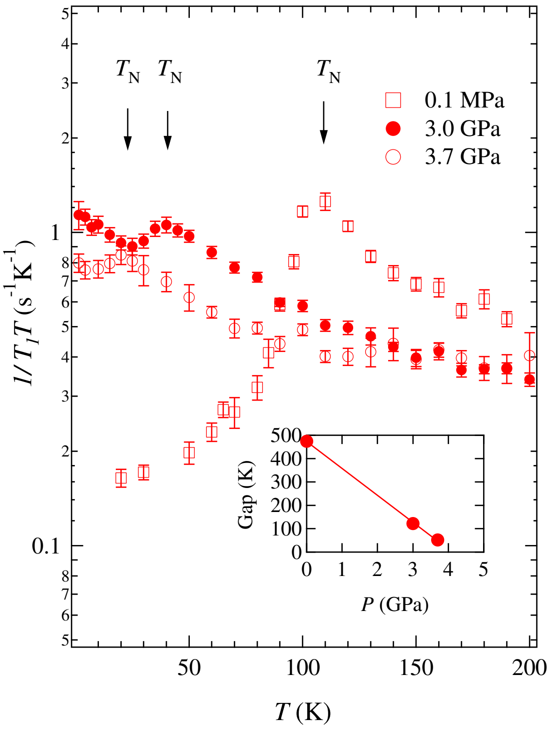

Figure 5: Relaxation rate of ^{75}As divided by temperature, $1/T_1T$ for $LaFeAsO_{0.4}H_{0.6}$. T_N represents the AF transition temperature. The inset shows the pressure dependence of the spin excitation gap Δ (See Eq. (1)).

shows $1/T_1T$ for ^{75}As, and the peaks correspond to T_N. The values of T_N determined from $1/T_1T$ are consistent with those obtained from the linewidth of ^{1}H. At low temperatures just below T_N, $1/T_1T$ is expressed as follows:

$$\frac{1}{T_1T} \propto e^{-\frac{\Delta}{T}} \qquad (1)$$

where Δ represent the spin excitation gap. The pressure dependence of Δ is shown in the inset to Fig. 5. Assuming that Δ shows the linear dependence, the pressure-induced QCP is estimated to be 4.1 GPa.

IV. Discussion

The activated spin excitation as shown in Eq. (1) originates from a spin density wave (SDW). However, $1/T_1T$ also shows Curie-Weiss behavior below

T_N. The behavior is not observed at ambient pressure and it is characteristic of the critical behavior near the pressure-induced QCP. The coexistence of the gapped and gapless excitations are specific to this system. In this system, major Fermi surfaces are electron pockets with a square-like shape in two dimensional k space. Some parts of the electron pockets would contribute to the nesting and the SDW formation. The critical behavior would originate from the other parts of the Fermi surfaces. The nesting condition becomes worse and the bandwidth becomes broader with increasing pressure. Owing to these effects, the activated behavior shown in Eq. (1) would disappear at the pressure-induced QCP.

V. Conclusions

We performed NMR measurements on $LaFeAsO_{0.4}H_{0.6}$ at 3.7 GPa to investigate the magnetic properties in the vicinity of the pressure-induced QCP. We have found that the SDW ordered state still remains at 3.7 GPa. The pressure-induced QCP is estimated to be 4.1 GPa from the pressure dependence of the spin excitation gap. The gapless excitation observed as the Curie-Weiss behavior of $1/T_1T$ coexists with the gapped excitation, implying that each excitation originates from different parts within the Fermi surfaces.

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