# Gapless magnetic excitation in a heavily electron-doped antiferromagnetic phase of $LaFeAsO_{0.5}D_{0.5}$

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Magnetic excitations in a heavily electron-doped antiferromagnet, $LaFeAsO_{0.5}D_{0.5}$, have been investigated using powder inelastic neutron scattering. Unlike other parent compounds of the iron-based superconductors, the magnetic excitation gap in $LaFeAsO_{0.5}D_{0.5}$ was not detected down to the lowest measured temperature of 4 K. This result can be understood as a result of quasi-isotropy within the $ab$ plane, which is consistent with the band calculation result that the $d_{xy}$ orbital plays the dominant role in the magnetism of $LaFeAsO_{0.5}H_{0.5}$. In addition, the intensities of the magnetic excitations in this phase are much stronger than those in nondoped LaFeAsO. Even in the paramagnetic phase, the magnetic excitation in $LaFeAsO_{0.5}D_{0.5}$ persists. These results corroborate recent studies showing that the electron doping enhances the localized nature in this system.

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## I. INTRODUCTION

Since the discovery of the first iron-based superconductor (FeSC), a wide variety of FeSC families have been discovered [1–3]. The different FeSC families commonly contain a conductive $FePn_4$ or $FeCh_4$ ($Pn$ = P, As; $Ch$ = Se, Te) layer and a blocking layer and have similar Fe 3$d$ electronic structures [3,4]. Typically, superconductivity emerges as a result of suppression of structural (orbital ordering) and magnetic phase transitions by carrier doping [2,3,5–10].

The highest class of a critical transition temperature, $T_c$, in bulk compounds has been found in a "1111" system, such as $Gd_{1-x}Th_xFeAsO$ [11] ($T_c$ = 56 K), $SmFeAsO_{1-\delta}$ [12], and $SmFeAsO_{1-x}F_x$ [13] ($T_c$ = 55 K). The $T_c$ for an FeSC tends to increase when the $FePn_4$ (or $FeCh_4$) tetrahedron forms a regular shape [14]. Indeed, the compounds listed above satisfy this empirical rule [14]. Intriguingly, however, the $FeAs_4$ tetrahedron in $LaFeAsO_{1-x}H_x$ deviates from a regular shape under an applied pressure of 6 GPa, despite its high $T_c$ of 52 K (for $x = 0.18$), which is comparable to the highest known value [15,16]. This result suggests that another factor plays a role in raising $T_c$ for the 1111 system besides the shape of the $FeAs_4$ tetrahedron.

Recently, a doping technique for producing a higher electron concentration for 1111 systems has been developed in which $H^-$ is used instead of $F^-$ [17–20]. Remarkably, owing to the heavy electron doping, a second antiferromagnetic phase appears in La- and Sm-1111 systems just after disappearance of the superconducting phase [21,22]. Several subsequent studies have all suggested that the heavy electron doping enhances electron correlations and the localized character in the second antiferromagnetic phase [8,19,20,23–25]. These studies suggested that spin and/or orbital fluctuations from the strongly correlated phase significantly contribute to the high $T_c$ in 1111 compounds [15,16,21,22]. Therefore, it is indispensable to understand magnetic excitations in the heavily electron-doped antiferromagnetic phase.

$LaFeAsO_{1-x}H_x$, a target material in this study, is the simplest 1111 system, since $La^{3+}$ does not have a magnetic moment. As shown in Fig. 1(a), LaFeAsO exhibits a structural transition with decreasing temperature from $P4/nmm$ (tetragonal) to $Cmme$ (orthorhombic) at $T_s$ = 155 K [26]. On further cooling, a magnetic phase transition occurs at $T_N$ = 137 K [26]. A magnetic structure in this phase (AF1 phase) is a collinear stripe type with a propagation vector $\boldsymbol{Q}_{AF1} = (1, 0, 1/2)$ in reciprocal lattice units (orthorhombic $Cmme$ unit cell) [26,27]. As shown in Fig. 1(b), the magnetic moments, which have a magnitude of 0.63 $\mu_B$/Fe, are parallel-antiparallel to the $a$ direction [26]. The magnetic ordering in the AF1 phase is compatible with a Fermi surface (FS) nesting scenario; it is regarded as a spin density wave (SDW) of itinerant electrons due to a FS nesting [5,28,29]. $LaFeAsO_{0.5}H_{0.5}$ also exhibits a magnetic phase transition at $T_N$ = 89 K subsequent to a tetragonal-orthorhombic structural transition at $T_s \sim 95$ K [21]. The space group changes from $P4/nmm$ to $Aem2$ during the structural transition. For comparison, throughout this paper we will employ the $Cmme$ type unit cell as in LaFeAsO to describe a magnetic structure in the heavily electron-doped antiferromagnetic parent phase (AF2 phase). The magnetic modulation vector, $\boldsymbol{Q}_{AF2}$, is (1,0,0), and the magnetic moments (1.21 $\mu_B$/Fe) are parallel-antiparallel

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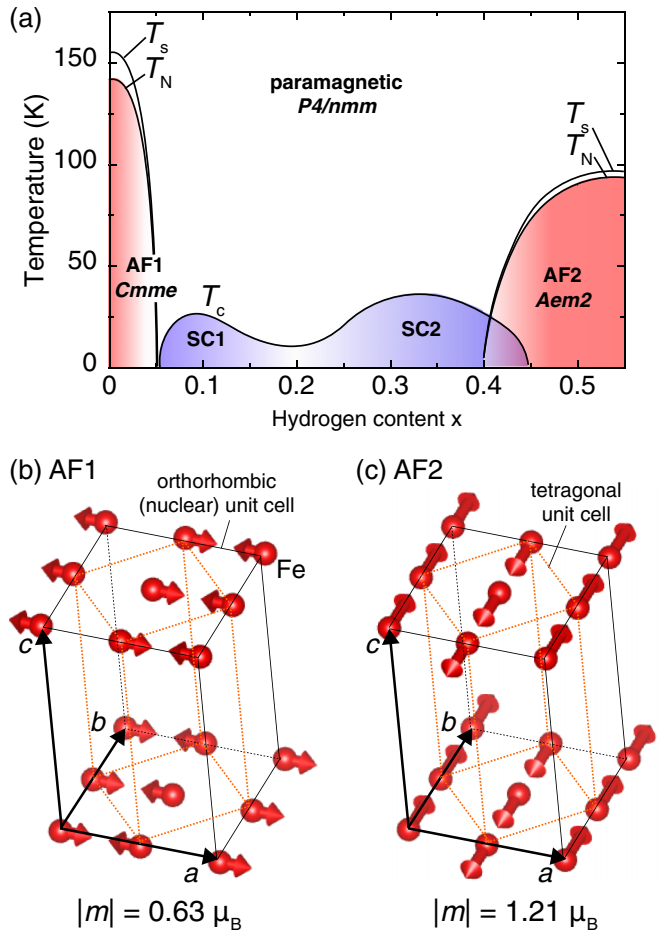


FIG. 1. (a) Schematic phase diagram of $LaFeAsO_{1-x}H_x$ as a function of hydrogen content. The target compounds in this work with $x = 0\,(x = 0.5)$ undergo structural transitions from $P4/nmm$ to $Cmme$ ($Aem2$) at $T_s = 155$ K (∼95 K), and then enter antiferromagnetic phases, AF1 (AF2), at $T_N = 137$ K (89 K) [21,26]. SC1 and SC2 represent the first and second superconducting phases with maximum critical transition temperature $T_{c,max} = 26$ K for $0.05 \leqslant x \leqslant 0.20$ and $T_{c,max} = 36$ K for $0.20 \leqslant x \leqslant 0.42$, respectively. Magnetic structures with their magnetic moments $m$ for (b) the AF1 phase and (c) the AF2 phase. For comparison, the orthorhombic $Cmme$ type unit cell (black solid lines) is used to describe both of the magnetic structures; the magnetic moments are parallel-antiparallel to the $a$ ($b$) direction in the AF1 (AF2) phases. A tetragonal unit cell above $T_s$ is also shown (orange dotted lines).

to the $b$ direction [see Fig. 1(c)] [21]. According to band calculations [19,20,24], the nesting is weakened by electron doping monotonically with $x$, and therefore, the magnetic ordering in the AF2 phase has little to do with the FS nesting. Thus, magnetic excitations in the AF2 phase are expected to exhibit features distinct from the typical SDW magnetism in LaFeAsO and in "122" families, such as $BaFe_2As_2$ [30–32].

In this paper, we report on magnetic excitations in the AF1 ($x = 0$) and AF2 ($x = 0.5$) phases, investigated through powder inelastic neutron scattering measurements on $LaFeAsO_{1-x}D_x$. We found that a spin gap in the AF2 phase, if present, is less than 0.6 meV. This observation is unique in the parent compounds of FeSCs. We suggest that the gapless excitation originates from quasimagnetic isotropy within the $ab$ plane. Moreover, we observed stronger magnetic excitations in the AF2 phase than those in the AF1 phase. The excitation in the AF2 phase can be observed even at 270 K. These results support the fact that the heavy electron doping enhances the localized nature in $LaFeAsO_{1-x}H_x$.

## II. EXPERIMENTS

Polycrystalline samples of nondoped LaFeAsO and deuterium-doped $LaFeAsO_{0.5}D_{0.5}$ were synthesized using solid-state reactions under ambient and high pressure, and they were accumulated to approximately 30 and 10 $g$, respectively [19,20,33]. To reduce the incoherent scattering contribution, deuterium was used instead of hydrogen. The inelastic neutron scattering measurements with an incident neutron energy $E_i$ of 150 meV were performed using the Fermi-chopper spectrometer, 4SEASONS, at BL01 of the Materials and Life Science Experimental Facility (MLF) in the Japan Proton Accelerator Research Complex (J-PARC) [34]. The Fermi-chopper frequency was set to 300 Hz, providing multiple incident energies of $E_i = 150$, 62.7, 34.2, 21.5, 14.8, 10.8, 8.2, and 6.4 meV [35]. The time-of-flight data from many detectors were converted to a $S(Q, E)$ map and analyzed using the UTSUSEMI software [36]. The lattice parameters used in our estimations of the absolute values of $|\boldsymbol{Q}_{\mathrm{AF1(2)}}|$ are $a = 5.71$ (5.62), $b = 5.68$ (5.60), and $c = 8.73$ (8.64) Å for $x = 0\,(x = 0.5)$, respectively [21,26].

## III. RESULTS AND DISCUSSION

Figure 2 shows the inelastic neutron scattering intensities of LaFeAsO and $LaFeAsO_{0.5}D_{0.5}$ as a function of the momentum transfer $Q$ and the energy transfer $E$ with an incident neutron energy of 21.5 meV. Strong intensities standing around $Q = 3.5$ Å$^{-1}$ are attributed to phonon scatterings from their temperature dependence [$\propto n(E) + 1$, where $n(E)$ is the Bose-Einstein factor] [37,38]. As indicated in Fig. 2(b), we observed weak magnetic scattering at $Q \sim 1.15$ Å$^{-1}$, which corresponds to the ordered momentum of AF1 with $|\boldsymbol{Q}_{\mathrm{AF1}}| = 1.16$ Å$^{-1}$, at 140 K ∼ $T_N$ for $x = 0$. At the lowest temperature, 4 K, the magnetic excitation disappears below 10 meV [Fig. 2(c)]. This indicates that an excitation gap of ∼10 meV opens at 4 K in the AF1 phase. Indeed, as the constant-energy cuts shown in Fig. 3(a) demonstrate, the magnetic excitation below 10 meV can be observed only at 140 K, while above 10 meV, it can also be seen at 4 K. These results are in good agreement with previous inelastic neutron scattering results [39,40].

At 90 K ∼ $T_N$ for $x = 0.5$ [Fig. 2(e)], a clear and rather broad magnetic scattering centered on $Q \sim 1.2$ Å$^{-1}$ was observed besides the phonon scattering around 3.5 Å$^{-1}$. This broad scattering consists of two magnetic excitations from $\boldsymbol{Q}_1 = \boldsymbol{Q}_{\mathrm{AF2}}$ and $\boldsymbol{Q}_2 = (1, 0, 1)$, where $|\boldsymbol{Q}_{\mathrm{AF2}}| = 1.12$ Å$^{-1}$ and $|\boldsymbol{Q}_2| = 1.33$ Å$^{-1}$ [see also Fig. 3(b)]. These $Q$ positions are consistent with the previous neutron diffraction study [21].

Compared with the results for LaFeAsO, there are two distinctive features in the magnetic excitation of $LaFeAsO_{0.5}D_{0.5}$. First, the magnetic excitation below 10 meV

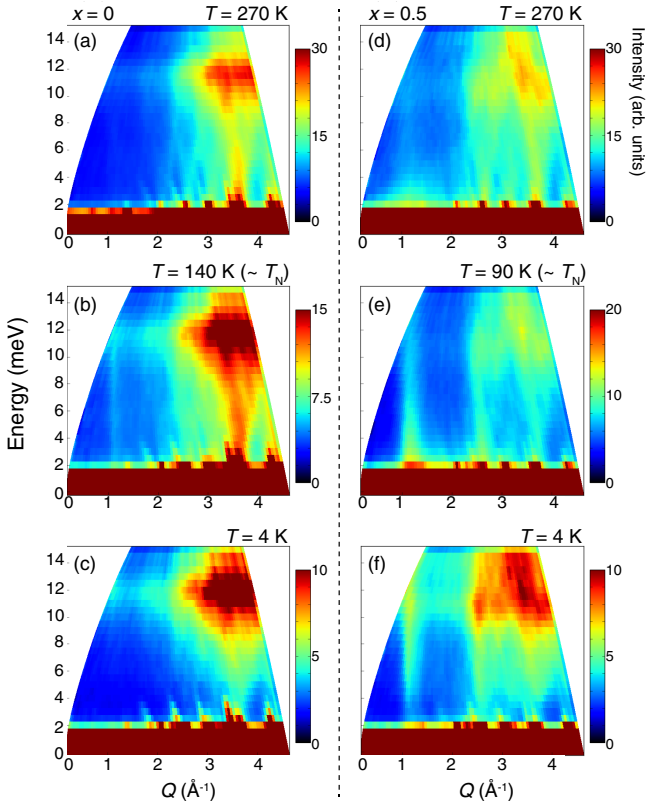


FIG. 2. Neutron scattering spectra of polycrystalline LaFeAsO [(a)–(c)] and $LaFeAsO_{0.5}D_{0.5}$ [(d)–(f)]. The data were recorded at 270 K [(a), (d)], and nearby $T_N$ (140 K for LaFeAsO and 90 K for $LaFeAsO_{0.5}D_{0.5}$) [(b), (e)], and 4 K [(c), (f)] with incident neutron energies of 21.5 meV. The intensities in this figure are normalized using incoherent elastic scattering intensities so as to enable comparison of the intensities for $x = 0$ with those for $x = 0.5$.

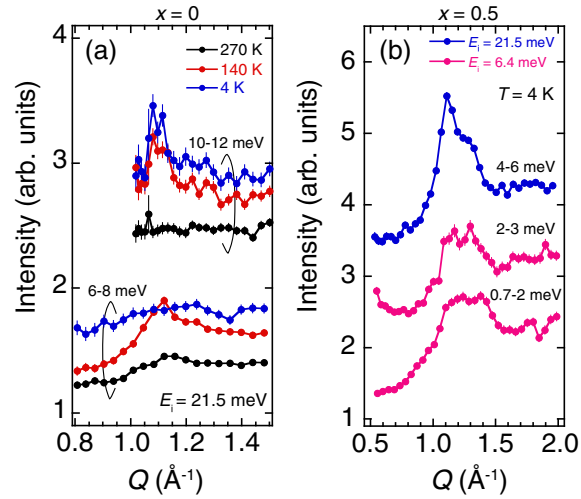


FIG. 3. (a) Constant-energy cuts showing the temperature dependence of the magnetic excitation at $Q \sim 1.15$ Å$^{-1}$ for $x = 0$ ($E_i = 21.5$ meV). The data were averaged over the indicated energy ranges and were divided by $n(E) + 1$ after constant background subtraction. Then, the data were vertically shifted by 1.5 for clarity. (b) Constant-energy cuts of the spectra at 4 K for $x = 0.5$. The data were averaged over the indicated energy ranges and were vertically shifted by 1 for clarity. The blue (pink) circles show the data for $E_i$ of 21.5 (6.4) meV, whose intensities are normalized by using incoherent elastic scattering intensities.

in $LaFeAsO_{0.5}D_{0.5}$ is clearly observed even at the lowest temperature, 4 K, as shown in Fig. 2(f). In addition, as evidenced by the results of the lowest $E_i$ of 6.4 meV in our measurements [Fig. 3(b)], the excitation gap (spin gap) in $LaFeAsO_{0.5}D_{0.5}$ is less than 0.6 meV (see the Supplemental Material [37]). This result is in striking contrast to the other parent compounds of FeSCs, in which the values of the excitation gap are usually $5 \sim 11$ MeV [37,39–45]. We will discuss this point in more detail later.

Second, as shown in Fig. 2(d), diffuse magnetic scattering can be observed on the same $Q$ position even at 270 K, which is comparable to $3T_N$ [46]. This indicates that there are persistent magnetic fluctuations above $T_N$ in $LaFeAsO_{0.5}D_{0.5}$. In the AF1 phase, a specific wave vector corresponding to the nesting vector, namely $\boldsymbol{Q}_{AF1}$, is favored at the magnetic transition. In contrast, because the nesting vector does not exist [19,24], several magnetic ordering states with different wave vectors compete with each other in $LaFeAsO_{0.5}D_{0.5}$. This condition should lower $T_N$ and leads to persistent magnetic fluctuations above $T_N$. Therefore, the paramagnetic scattering observed in $LaFeAsO_{0.5}D_{0.5}$ shows that the magnetic ordering in the AF2 phase occurs without the FS nesting. The smaller $T_N$ for $x = 0.5$, despite the larger moment, should be a reflection of the enhanced magnetic fluctuations.

Figures 4(a) and 4(b) show the energy dependence of the imaginary part of the dynamic magnetic susceptibility, $\chi''(Q_{AF1(2)}, E)$. The temperature variations of $\chi''(Q_{AF1(2)}, E)$ in the AF1 (AF2) phase were derived as follows: After background subtraction, the energy dependence of the averaged intensities at $Q = 1.10$–$1.20$ ($1.07$–$1.17$) Å$^{-1}$ were divided by $n(E) + 1$, and the backgrounds were estimated as the averaged intensities at $Q = 0.80$–$0.90$ and $1.50$–$1.60$ Å$^{-1}$. The data for $x = 0$ are consistent with results of a previous study [39]. The intensity of the AF2 magnetic scattering is about four times stronger than that of the AF1 magnetic scattering at their respective values of $T_N$. Since the magnetic reflection detected by the neutron scattering technique is proportional to the square of the magnitude of the ordered magnetic moment, this result is consistent with the magnitude of the ordered magnetic moments, $0.63\,\mu_B$ for LaFeAsO [26] and $1.21\,\mu_B$ for $LaFeAsO_{0.5}D_{0.5}$ [21].

To date, inelastic neutron scattering studies of magnetic excitations in the parent compounds of FeSCs have been extensively conducted [31,32]. These studies revealed that the spin gap is commonly observed in the parent compounds of FeSCs, which include both metallic and insulating systems [37]. Under these circumstances, the almost gapless excitation in the AF2 phase is remarkably unique.

We now suggest that the gapless excitation in the AF2 phase originates from the lack of anisotropy within the $ab$ plane. As Moon *et al.* demonstrated [24], for $x = 0$, the $d_{yz}$ and $d_{xy}$ orbitals have a spin polarization in their spectral function, while the spin polarization of the $d_{xz}$ orbitals is smaller. This difference in the spin polarization between the $d_{xz}$ and $d_{yz}$ orbitals should produce anisotropy within the $ab$

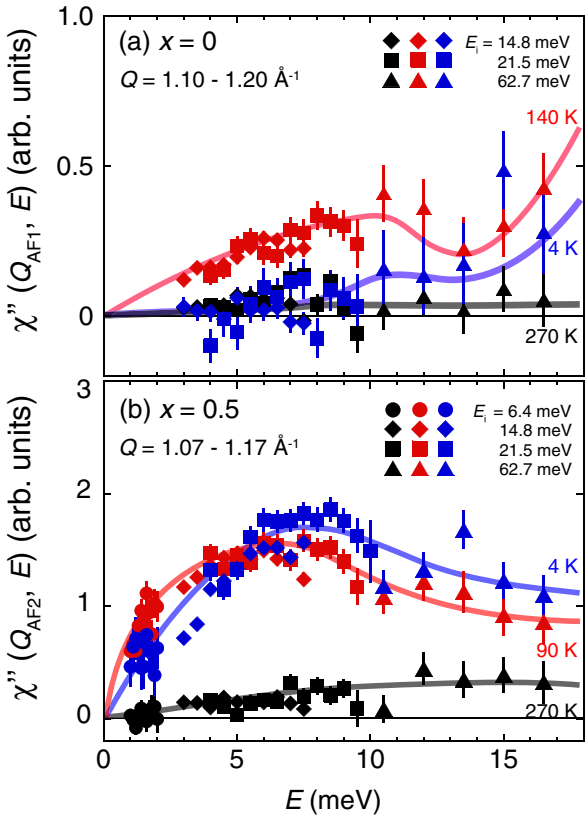


FIG. 4. Energy dependence of the imaginary part of the dynamic magnetic susceptibility, $\chi''(Q_{\mathrm{AF1(2)}}, E)$ at each temperature for (a) $x = 0$ and (b) $x = 0.5$. Note that the $\chi''(Q_{\mathrm{AF1}}, E)$ with $E_i = 6.4$ meV could not be measured with a sufficient signal-to-noise ratio. The intensities for different $E_i$'s and $x$'s are normalized by using incoherent elastic scattering intensities so as to make the comparison easy. Solid lines are guides for the eyes.

plane. On the other hand, for $x = 0.5$, the electron doping makes the $d_{xy}$ orbital the most dominant component of the static magnetic moments in the AF2 phase [24]. In contrast, the $d_{xz/yz}$ orbitals have no longer significant spin polarization. In addition, it is pointed out that the large magnetic moment in the AF2 phase is due to the enhanced effective Coulomb repulsion and the reduced hopping in the $3d_{xy}$ orbital that results from electron doping [8,20]. This situation would result in negligible anisotropy within the $ab$ plane. As a result, the almost gapless excitation in the AF2 phase is realized [47]. To our knowledge, this is the first reported case of gapless magnetic excitation in the commensurate phases of the parent compounds of FeSCs, except for that in the incommensurate magnetic phase of $Fe_{1.141}Te$ [48]. This is by virtue of the heavy electron-doping technique using $H^-$, which leads to the $d_{xy}$-dominant magnetic state.

Finally, based on the fact that the magnetic excitation in the AF2 phase clearly reflects the $d_{xy}$-dominant magnetic properties, we suggest that magnetic fluctuations within the $ab$ plane also play a significant role in the emergence of the SC2 phase. This situation seems to resemble cuprate superconductors, in which a single $d_{x^2-y^2}$ orbital governs the superconductivity [49].

## IV. SUMMARY

In summary, we investigated magnetic excitations in the AF2 phase of $LaFeAsO_{0.5}D_{0.5}$ using powder inelastic neutron scattering. We found that the excitation gap in the AF2 phase is less than 0.6 meV, which is in striking contrast to other parent compounds of FeSCs. Based on the $d_{xy}$-dominant state specific to $LaFeAsO_{0.5}H_{0.5}$, we suggest that the gapless excitation is due to a lack of anisotropy within the $ab$ plane. In addition, the magnetic excitation in $LaFeAsO_{0.5}D_{0.5}$ is observed even in the paramagnetic phase, and the intensities of the magnetic excitation in the AF2 phase are much stronger than those in the AF1 phase. These results suggest the more localized nature in the AF2 phase than that in the AF1 phase. This study demonstrates a typical case of magnetic excitation in a parent compound of an FeSC where the $d_{xy}$ orbitals play the most dominant role in magnetism.

## ACKNOWLEDGMENTS

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[46] Note that although such a paramagnetic scattering was also observed in the other parent compounds, such as $AFe_2As_2$ ($A$ = Ba, Ca) [51,52], NaFeAs [53], and $Fe_{1.1}Te$ [54], its property appears to be distinct from family to family.

[47] From the standpoint of the localized Heisenberg picture, this scenario can be interpreted as follows: A spin Hamiltonian for $x = 0$ should contain both in-plane and out-of-plane anisotropy constants, while for $x = 0.5$, the in-plane anisotropy constant is zero. Although the out-of-plane anisotropy would still exist in $LaFeAsO_{0.5}H_{0.5}$, its value (sign) is considered to be in favor of the spin arrangement lying within the $ab$ plane, as in $x = 0$ [40]. Because a powder sample is used in our measurements, an excitation gap arising from the out-of-plane anisotropy should be overlapped by the gapless magnetic excitation observed in our measurements.

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