

## NMR evidence of strongly correlated superconductivity in LiFeAs: Tuning toward a spin-density-wave ordering

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In this Rapid Communication, we reported the results of NMR study on LiFeAs single crystals. We find a strong evidence of the low-temperature spin fluctuations; by changing sample preparation conditions, the system can be tuned toward a spin-density-wave (SDW) quantum-critical point. The detection of an interstitial Li(2) ion, possibly locating in the tetrahedral hole, suggests that the off-stoichiometry and/or lattice defect can probably account for the absence of the SDW ordering in LiFeAs. These facts show that LiFeAs is a strongly correlated system and the superconductivity is likely originated from the SDW fluctuations.

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The interplay of magnetism and superconductivity is one of the dominant themes in the study of unconventional superconductors, such as high- $T_c$  cuprates, organic superconductors, and heavy fermions, where the magnetic fluctuations are crucial to the superconductivity, in general.<sup>1-3</sup> This subject has also been extensively studied both experimentally and theoretically in the recent discovered iron pnictides,<sup>4</sup> where high-temperature superconductivity is achieved by suppressing a competing spin-density-wave (SDW) state upon chemical doping or pressure<sup>5-7</sup>. Here the superconductivity emerging in proximity to a SDW quantum-critical point, as well as the persisting spin fluctuations shown above  $T_C$ ,<sup>8</sup> support strongly that superconducting pairing is mediated by spin fluctuations.

However, in an iron pnictide LiFeAs, bulk superconductivity up to 18 K, instead of long-range antiferromagnetism (AFM), is found in the ground state without nominal doping.<sup>9-11</sup> Angle-resolved photoemission spectroscopy studies<sup>12-14</sup> do not see evidence of spin fluctuations. In particular, the superconducting gap seems to be a single isotropic gap with a moderate amplitude,<sup>12-14</sup> in contrast to the multiple gaps in other iron pnictides which is likely originated from Fermi-surface nesting and spin fluctuations.<sup>15-20</sup> The  $\mu$ SR studies show that LiFeAs has a different Uemura relation with other pnictide superconductors.<sup>21</sup> These facts lead to an everlasting proposal that LiFeAs is a conventional superconductor, rather than a strongly correlated superconductor.

Theoretically, the local-density approximation calculations indicate that LiFeAs has a similar band structure with LaFeAsO, BaFe<sub>2</sub>As<sub>2</sub>, and NaFeAs, and therefore, a similar magnetic ordering and a universal origin of superconductivity are expected among all compounds.<sup>22-25</sup> Particularly, the sister compound NaFeAs with the same 111 structure has similar magnetic<sup>26,27</sup> and superconducting properties<sup>28</sup> (upon doping) to the 1111 [such as RFeAsO<sub>1-x</sub>F<sub>x</sub> (Refs. 5, 29, and 30)] and the 122 (such as Ba<sub>1-x</sub>K<sub>x</sub>Fe<sub>2</sub>As<sub>2</sub> and Ba(Fe<sub>1-x</sub>Co<sub>x</sub>)<sub>2</sub>As<sub>2</sub> (Refs. 6 and 31)] families. It is conjectured that the absence of AFM in LiFeAs is probably caused by lithium deficiency.<sup>10</sup> However, the lacking of evidences for chemical nonstoichiometry and the absence of Curie-Weiss-type low-temperature SDW spin fluctuations from

NMR (Refs. 32 and 33) do not seem to support this scenario.

Therefore, the study of whether LiFeAs is a strongly correlated superconductor is certainly important for understanding the correlation among the band structure, the magnetism, and the mechanism of superconductivity of the high  $T_c$  pnictides. In order to resolve this problem, we performed NMR studies on LiFeAs single crystals. We first searched for possible SDW order and SDW fluctuations in our high-quality single crystals. We found evidences of anisotropic spin fluctuations, which can be tuned toward an SDW quantum-critical point. We further show spectral evidence that the absence of antiferromagnetism in LiFeAs is likely caused by the doping and/or the scattering effect from an additional Li(2) site.

The single crystals of LiFeAs were grown by self-flux method with two different growth conditions. LiAs was first synthesized as precursor by reacting Li (3N) and As (5N) in Ta tube sealed in evacuated quartz tube and heated at 600 °C for 10 h. Mixtures of LiAs and Fe (4N) powder with the composition of Li<sub>5</sub>FeAs<sub>5</sub> were sealed into Ta tubes under 1.5 atm of argon gas, then the Ta tubes were vacuum sealed into quartz tubes. For the sample 1 (S1), the tube was heated to 1050 °C, held for 24 h and cooled slowly to 650 °C over 200 h, then the furnace is shut down and naturally cooled down to room temperature. The sample 2 (S2) was obtained by heating the tube to 1170 °C, held there for 2 h before the temperature was decreased to 870 °C within 100 h, then the sample was taken out and quenched in air. The S1 has a large superconducting volume and a higher  $T_c$  17 K, whereas the S2 has a smaller superconducting volume and a lower  $T_c$  10 K [see Fig. 1(b)]. Their electronic and magnetic properties were measured on a quantum design physical property measurement system with the vibrating-sample magnetometer option provided.

The NMR crystals were chosen with typical dimension of  $3 \times 2 \times 0.1$  mm<sup>3</sup>. Both <sup>7</sup>Li ( $S=3/2$ ) and <sup>75</sup>As ( $S=3/2$ ) NMR studies were performed with the magnetic field along the  $ab$  plane and the  $c$  axis. All spectral measurements use the spin-echo technique. The spin-lattice relaxation rate is deduced from an inversion-recovery method and the spin magnetization is fit with the  $S=3/2$  nuclear recovery  $\frac{m(t)}{m(0)} = 1 - A(0.1e^{-t/T_1} + 0.9e^{-6t/T_1})$  for <sup>75</sup>As and  $\frac{m(t)}{m(0)} = 1 - A(0.1e^{-t/T_1} + 0.9e^{-6t/T_1})$  for <sup>7</sup>Li.

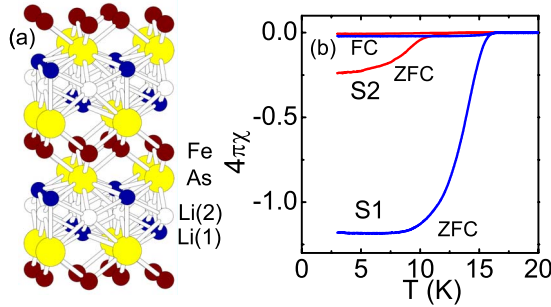


FIG. 1. (Color online) (a) The proposed crystal structure of  $\text{Li}_{1+x}\text{FeAs}$  (Refs. 10 and 34). Additional Lithium ion Li(2) (hollow circle) occupies the interstitial site (3/4, 1/4, and 1/2), which is right above the Fe site and inside an  $\text{As}_4$  tetrahedron. (b) The dc susceptibility of two LiFeAs crystals S1 and S2 with different growth conditions with zero-field-cooled and field-cooled conditions under 100 Oe field.

We first study the spin fluctuations through the spin-lattice relaxation rate  $1/T_1$  of  $^{75}\text{As}$  in LiFeAs. In Fig. 2(a), the  $1/^{75}T_1^{ab}T$  of the superconducting crystal S1 ( $T_c \approx 17$  K) is shown with an 8 T magnetic field along the  $ab$  plane. The superconducting onset is shown by a sharp drop of the  $1/^{75}T_1T$  below  $T_c$ . There is a small upturn while temperature decrease from 50 K down to  $T_c$ , which indicate spin fluctuations. Following Moriya's two-dimensional spin-fluctuation theory in a paramagnet, we fit the data with a Curie-Weiss behavior  $1/T_1T = A/(T + \Theta) + b$  at low temperatures [see Fig. 2(a)]. Here  $A$  is proportional to the electron density of state on the Fermi surface, and the value of  $\Theta$  is correlated with band mass  $m^*$ , whose sign usually switches from negative to positive if the system is tuned from an antiferromagnetic ground state to a quantum-disordered paramagnet. The  $b$  term is obtained phenomenologically assuming a Korringa (Fermi liquid) contribution from the multiple band system. Our fitting parameter  $\Theta = 30 \pm 5$  K is comparable with the

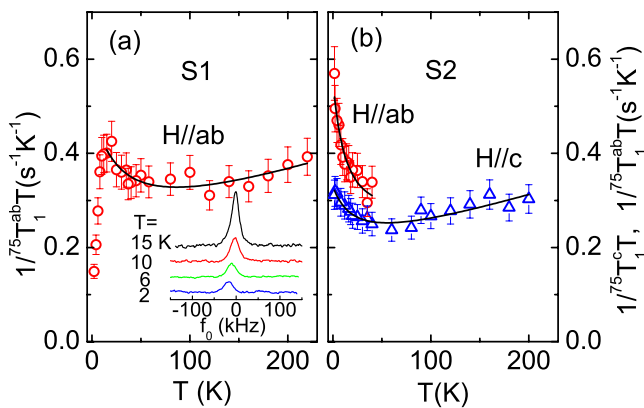


FIG. 2. (Color online) (a) The spin-lattice relaxation rate  $1/^{75}T_1T$  of sample S1 under a field of 8 T applied along the crystal  $ab$  plane. The solid line is a fit by a Curie-Weiss term  $1/T_1T = A/(T + \Theta) + b + cT$  with  $\Theta = 30 \pm 5$  K. The  $^{75}\text{As}$  NMR spectra at different temperatures are shown. (b) The  $1/^{75}T_1T$  of sample S2 with field applied along  $ab$  plane and the  $c$  axis. The solid line is a guide to the Curie-Weiss fitting with  $\Theta = 10 \pm 5$  K for field along the  $ab$  plane and  $\Theta = 20 \pm 5$  K for field along the  $c$  axis.

optimal-doped  $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ ,<sup>8</sup> indicating a similar strength of spin fluctuations with other iron pnictide superconductors.

To check the sample dependence of spin fluctuations, we investigated the spin-lattice relaxation of sample S2 with a less superconducting volume and a lower  $T_c$ . The spin-lattice relaxation rate (SLRR) of the crystal S2 is measured with field applied both along the  $ab$  plane ( $1/^{75}T_1^{ab}T$ ) and along the  $c$  axis ( $1/^{75}T_1^cT$ ) [see Fig. 2(b)]. Comparing with S1, the low temperature  $1/^{75}T_1^{ab}T$  of S2 are very different. The  $1/^{75}T_1^{ab}T$  increase dramatically as temperature drops under a 12 T magnetic field with  $\Theta \approx 10 \pm 5$  K, which is close to a diverging behavior (i.e.,  $\Theta \leq 0$  K) at finite temperature. Such behavior is a clear indication that the system is close to a magnetic ordering.

Next we discuss the nature of the low-temperature spin fluctuations. As shown in Fig. 2, the Curie-Weiss behavior is also seen in  $1/^{75}T_1^cT$  ( $\Theta \approx 20 \pm 5$  K) but much weaker than that of the  $1/^{75}T_1^{ab}T$ . The anisotropy of the spin-lattice relaxation rate defined as  $T_1^c/T_1^{ab}$ , increases as temperature drops, which is well described as signatures of the stripe AFM (or the SDW) correlations.<sup>35-37</sup> The observations of the dramatic enhancement and the anisotropy of the low-temperature spin-lattice relaxations indicate unambiguously that LiFeAs is a strong-correlated system, which is close to the SDW order.

This draws a similarity among LiFeAs and other iron pnictides and suggests that the superconductivity in LiFeAs is also mediated by SDW spin fluctuations. Although the long-range magnetic ordering is not achieved so far in our crystals, the small value of  $\Theta$  in S2 indicates that the LiFeAs is tuned toward an SDW quantum-critical point. Obviously it is important to find what the tuning parameter is. The sample difference is probably related to the chemical off-stoichiometry which contribute to a doping effect and/or an impurity scattering effect.

In fact, it is well-known that lithium could be reversibly intercalated/inserted into the interstitial sites for many materials due to its high mobility and small size such as in lithium ion batteries. Considering such behavior of lithium and the structure of LiFeAs, we performed  $^7\text{Li}$  spectrum studies to search for possible extra Lithium in the lattice. With the magnetic field along the  $ab$  plane, we found a very weak spectral tail on the low-frequency side of the normal Li spectrum in S1 [see Fig. 3(a)] and we label two subspecies as Li(1) and Li(2). Their respective spin-lattice relaxation rates,  $1/^{7}T_1(1)$  and  $1/^{7}T_1(2)$ , are also compared [see Fig. 3(b)]. A weak upturnlike behavior is seen in both  $1/^{7}T_1T$  as temperature decreases, which is probably also caused by the spin fluctuations as shown in  $1/^{75}T_1T$ . The Li(1) [one central transition and two satellites with  $^7\nu_q(1) \sim 0.06$  MHz] is identified as normal lithium site as shown in Fig. 1 because of its large intensity (multiplied by 0.1 in the figure), low hyperfine coupling from its small Knight shift ( $^7K_n \ll 0.005\%$ ) and slow spin-lattice relaxation rate. The Li(2) is on another specific site because its spin-lattice relaxation follows a single  $T_1$  component (data not shown). Both Li(1) and Li(2) spectra broaden below  $T_c$ , which leads to more spectral overlap in the frequency range. In order to reduce the overlap, we use a fast recovery method, where the samples are fed with saturation pulses and then the measurements are

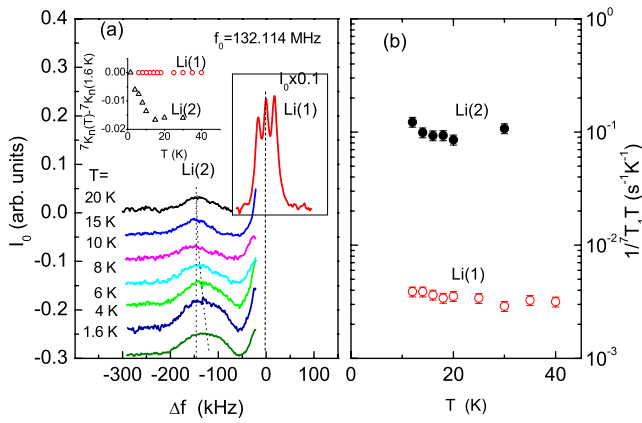


FIG. 3. (Color online) (a) The  ${}^7\text{Li}$  NMR spectrum of a LiFeAs crystal (S1) with 7.98 T field in the  $ab$  plane. The spectrum splits into two species, Li(1) and Li(2). The Li(2) spectrum below  $T_c$  is obtained by a fast recovery method (see text); inset: the Knight shift of Li(1) and Li(2) (relative to  $T=1.6$  K). (b) The comparison of the SLRR between Li(1) and Li(2). The  $1/T_1$  on the Li(2) site is 30 time faster but in the same order with  $1/{}^{75}\text{As}$  of  ${}^{75}\text{As}$ .

taken after a short recovery time  $t[t \approx \frac{1}{6}{}^7T_1(2) \ll {}^7T_1(1)]$ . Since Li(1) has a much longer  $T_1$  than that of Li(2) [see Fig. 3(b)], the spectral weight contribution of Li(1) at the Li(2) frequency is then negligible.

The Li(2) spectra shift to higher frequency [see Fig. 3(a) inset] and broaden right below  $T_c$ , which indicates that Li(2) is intrinsic in a superconducting state. We are not able to find the satellite of Li(2) and assign the lattice position of Li(2) directly, possibly because the satellite intensity of Li(2) is too low. According to the spectrum and the spin-lattice relaxation measurements, it is natural to conclude that the Li(2) comes from an interstitial site in the lattice [see Fig. 1(a)], which is located just above the Fe site and enclosed in an  $\text{As}_4$  tetrahedron.<sup>34</sup> Since Li has a very small quadrupole moment, the quadrupole correction to the center frequency of Li(2) is probably negligible.<sup>38</sup> The Knight shift of Li(2) with field along the  $ab$  plane is estimated with  ${}^7K_n^{ab} \sim -0.1\%$  much larger than that of Li(1). Li(2) is also found with field along the  $c$  axis and Knight shift is estimated as  ${}^7K_n^c \sim -0.05\%$ . Early x-ray studies<sup>34</sup> on LiFeAs suggest an interstitial Li site located above the Fe site and enclosed in an  $\text{As}_4$  tetrahedron, which seems consistent with our Li(2) data for several reasons: (1) a large hyperfine coupling of the Li(2) to the Fe is expected unlike the cancellation effect of the diagonal hyperfine field on the Li(1) position.<sup>27,35</sup> This is consistent with the fast spin-lattice relaxation rate and a large Knight shift of Li(2). (2) The hyperfine coupling  $A_{hf}^c$  is expected stronger than  $A_{hf}^{ab}$ , which is consistent with the anisotropic Knight shift of Li(2). (3)  ${}^{57}\text{Fe}$  also has a negative Knight shift from spin contributions with  ${}^{57}K_n \sim -0.1\%$ ,<sup>39</sup> which supports that Li(2) is located at the same symmetric position as iron.

It should be noted that, in  $\text{Li}_{1+x}\text{MnAs}$ ,<sup>40</sup> the (3/4, 1/4, and 1/2) sites [see Fig. 1(a)] are fully occupied by lithium, whereas only a few percent of additional lithium is located in octahedral holes. One can hence speculate that lithium deficiency and/or insertion can occur simultaneously in

$\text{Li}_{1+x}\text{FeAs}$ . We roughly estimate the Li(2) concentration of  $(6 \pm 3)\%$ /unit in S1, by comparing the spectral weight between Li(1) and Li(2) and assuming Li(2) satellites do not overlap with the center line [otherwise  $(2 \pm 1)\%$ /unit]. Compared with the sister 111 compound NaFeAs,<sup>28</sup> such a carrier level is sufficient to tune the SDW and superconducting state.

Now we discuss the cause of property difference between sample S1 and S2. Li(2) is also found in sample S2, estimated with a concentration of  $(9 \pm 3)\%$ /unit [or  $(3 \pm 1)\%$ /unit with overlapping center line and satellites]. However, it is unreliable to compare the concentration difference within the error and we consider several possibilities. If the actual concentration of Li(2) is higher in S1, a larger electron-doping effect is expected to suppress the SDW and induces the superconductivity, which is seen in S1. On the other hand, if the actual concentration of Li(2) is higher in S2, additional doping effect such as Li(1) deficiency, may exist to cancel the Li(2) effect. Then a higher concentration of Li(2) in S2 result in the low  $T_c$  and bring the system back toward the SDW ordering.<sup>41</sup> In fact, lithium deficiency on Li(1) has been speculated to coexist with Li(2) doping.<sup>10,22,40</sup> Besides, disorder could also play an essential role here. Since S2 is made with a fast growth condition, disorder scattering may suppress the superconductivity and favor the competing SDW ordering. Finally, we should also point out that although Li(2) is coupled to superconductivity, we cannot completely rule out the possibility that the Li(2) is from a minor superconducting phase. More work is needed to verify the position and the role of Li(2) proposed in LiFeAs.

To summarize, we found two independent evidences for strong-correlated superconductivity in LiFeAs. First, evidence of strong spin fluctuations is found in the normal state right above  $T_c$ , which increase as temperature drops. Such effect supports that superconductivity is probably mediated by spin fluctuations. In particular, a suppression of  $T_c$  with different growth conditions leads to a significant enhancement of anisotropic spin fluctuations toward an SDW quantum-critical point. Second, our data show a Li(2) signal with a finite concentration in superconducting LiFeAs and the absence of the long-range AFM in LiFeAs could be caused by a doping effect. Combining both evidences, our data unifies LiFeAs with the 1111 and the 122 iron pnictides with the same magnetic origin and the same mechanism of superconductivity. We believe that our results are important to understand the mechanism of superconductivity and underline further the importance of magnetic fluctuations for the superconductivity pairing observed in iron-based superconductors.

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