High-\(T_c\) Nodeless \(s_\lambda\)-wave Superconductivity in \((Y,La)FeAsO_{1-y}\) with \(T_c=50\) K: \(^{75}\text{As}-\text{NMR}\) Study

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We report \(^{75}\text{As}-\text{NMR}\) study on the Fe-pnictide high-\(T_c\) superconductor \(Y_{0.95}\text{La}_{0.05}\text{FeAsO}_{1-y}\) \((Y_{0.95}\text{La}_{0.05}1111)\) with \(T_c=50\) K that includes no magnetic rare-earth elements. The measurement of the nuclear-spin lattice-relaxation rate \(^{75}(1/T_1)\) has revealed that the nodeless bulk superconductivity takes place at \(T_c=50\) K while antiferromagnetic spin fluctuations develop moderately in the normal state. These features are consistently described by the multiple fully-gapped \(s_\lambda\)-wave model based on the Fermi-surface (FS) nesting. Incorporating the theory based on band calculations, we propose that the reason that \(T_c=50\) K in \(Y_{0.95}\text{La}_{0.05}1111\) is larger than \(T_c=28\) K in \(La1111\) is that the FS multiplicity is maximized, and hence the FS nesting condition is better than that in \(La1111\).

After the discovery of superconductivity (SC) in iron (Fe)-based oxypnictide \(\text{LaFeAsO}_{1-x}\) \((\text{LaFeAsO}_{1-x}F_x\) (hereafter denoted as \(\text{La1111}\)) with a SC transition temperature \(T_c=26\) K\(^3\), \(T_c\) goes up over 50 K with the replacement of \(\text{La}\) for other magnetic rare-earth elements in \(\text{Ln1111}\) \((Ln=\text{Sm},\text{Nd} \ldots)\)\(^2\,4\). Over the past four years, extensive studies have been reported on various Fe-based superconductors, pointing to the diversity of SC characteristics and normal-state electronic properties. It is believed that this diversity is associated with the multiband/multiorbital nature of the degenerate Fe-3d states in the Fe\(Pn\)\((Pn=\text{As},\text{P})\) layer. In the lattice-parameter points of view, \(T_c\) reaches a maximum of 55 K when a Fe\(As_d\) block forms in a nearly regular tetrahedral structure in \(\text{Ln1111}\)\(^5\), where the height of pnictogen\((h_{\text{p}})\) from the Fe plane and the \(a\)-axis length are \(h_{\text{p}}\approx1.38\AA\)\(^6\) and \(a\approx3.9\AA\)\(^7\,8\), respectively. In order to address a possible mechanism for high-\(T_c\) SC in \(\text{Ln1111}\), it is desired to gain further insight into why a structure of \(\text{FeAs}_4\) tetrahedron is relevant with the diversity of SC characteristics and normal-state electronic properties. However, most \(\text{Ln1111}\) are not extensively investigated by various measurements of angle-resolved-photoemission spectroscopy (ARPES), NMR, scanning tunneling spectroscopy (STS), and so on. This partly because high-quality single crystals with a sufficiently large size are not yet available. The presence of \(4f\)-electrons derived magnetic fluctuations in \(\text{Ln1111}\) has prevented us from characterizing SC and normal-state properties by means of NMR measurements\(^9\,11\).

In this Letter, we report for the first time a \(^{75}\text{As}-\text{NMR}\) study on a high-\(T_c=50\) K-class of Fe-pnictide superconductor \(Y_{0.95}\text{La}_{0.05}\text{FeAsO}_{1-y}\) \((Y_{0.95}\text{La}_{0.05}1111)\) that does not involve any magnetic rare earth ions. We demonstrate that antiferromagnetic spin fluctuations (AFSFs) in \(Y_{0.95}\text{La}_{0.05}1111\) are more significant due to the better Fermi-surface (FS) nesting condition than those in \(\text{La1111}\) with \(T_c=28\) K, and as a result, \(Y_{0.95}\text{La}_{0.05}1111\) realizes the multiple fully gapped high-\(T_c\) \(s_\lambda\)-wave SC with \(T_c=50\) K.

A polycrystalline sample of \(Y_{0.95}\text{La}_{0.05}\text{FeAsO}_{1-y}\) \((\text{Ho}_{15}\) was synthesized via a high-pressure synthesis technique with an addition of a small amount of hydrogen as a sort of catalyst to stabilize homogeneous samples\(^3\,7\,12\). The nominal oxygen content is \(y=0.2 \sim 0.25\), but the actual oxygen content is slightly smaller than the nominal one, owing to the oxidation of starting rare-earth elements. The x-ray diffraction measurement indicates that the sample is composed of almost only a single phase with lattice parameters of \(a=3.863\AA\) and \(c=8.337\AA\), although a tiny amount of unreacted \(Y\)As is identified. As shown in Fig.\(^1\)(a), the \(a\)-axis length is compatible to that of \(\text{La1111}\) when \(T_c \geq 50\) K\(^4\,7\,8\). Bulk SC with \(T_c=50\) K for \(Y_{0.95}\text{La}_{0.05}1111\) was determined from an onset of SC diamagnetism in susceptibility (see Fig.\(^1\)(c)). A coarse powder sample was used for the measurements of nuclear spin-lattice relaxation rate \(^{75}(1/T_1)\) of \(^{75}\text{As}-\text{NMR}\) at the field \(\mu_B H \sim 12\) T perpendicular to the \(c\)-axis. Note that the \(^{75}\text{As}-\text{NMR}\) spectrum in \(Y_{0.95}\text{La}_{0.05}1111\) was discriminated from that of unreacted YAs.

Figure\(^1\)(d) and its inset show respective \(^{75}\text{As}-\text{NMR}\) and NQR spectra for \(Y_{0.95}\text{La}_{0.05}1111\). The estimated \(^{75}\text{As}-\text{NQR}\) frequency \(^{75}\nu_{Q}\) is 17.1 MHz, which is larger than \(^{75}\nu_{Q}\) of \(\text{La1111}\)\(^17\). The data of \(^{4}\text{FeAs}_{4}\) \((\text{FeAs}_{4}\text{M}_{2}\text{O}_{8}\) \(M=\text{Li,Na}\) \((\text{A111})\)\(^13\,14\), and \(^{2}\text{FeAs}(\text{A}=\text{Li,Na})\) \((A111)\)\(^15\,16\), as drawn by the dashed line in Fig.\(^1\)(b). Note that as the \(a\)-axis length decreases, \(^{75}\nu_{Q}\) increases linearly. Since \(^{75}\nu_{Q}\) in proportion to an electric field gradient at As nuclear site is determined by some charge distribution around the \(^{75}\text{As}\) nucleus in the \(\text{FeAs}_4\) tetrahedron, the increase in \(h_{\text{p}}\) in association with the reduction in \(a\)-axis increases \(^{75}\nu_{Q}\), yielding a monotonous variation in the covalency of Fe-As bond\(^8\).

Figure\(^2\)(a) shows the \(T\) dependence of \(^{75}\nu_{Q}\)\(^{1}/T_1 T\) (solid squares) normalized by the value at \(T=250\) K.
and 0. In general, \( K \) \cite{17, 19}, and La\(_{1111}\)(HOVD) \cite{75}, with Ba \( \chi \) dependent hyperfine-coupling constant and 0 as \( Y \) \( \chi \) increases from 28 K to 34 K and then to 50 K at \( T \sim 250 \) K. A sharp NMR spectrum denoted by \( * \) comes from a small amount of unreacted YA sample, which is separately discriminated. Inset: \( ^{75} \)As-NQR spectra of \( Y_{0.95}\)La\(_{0.05}\)1111, La\(_{1111}\)(H)\(|\text{La}(H)|\sim\text{32 K}\) \cite{15, 16}, and La\(_{1111}\)(\( T_c \approx 28 \) K) \cite{17}.

for \( Y_{0.95}\)La\(_{0.05}\)1111 and other La-based compounds such as \( Y_{0.92}\)La\(_{0.8}\)1111 \( (T_c = 34 \) K) \cite{18}, La\(_{1111}\)(OPT) \( (T_c = 28 \) K) \cite{17, 19}, and La\(_{1111}\)(HOVD) \( (T_c = 5 \) K) \cite{20}, along with Ba\(_{0.6}\)K\(_{0.4}\)Fe\(_2\)As\(_2\) (BaK122) \( (T_c = 38 \) K) \cite{19} and (Cu\(_4\)Al\(_2\)O\(_6\))Fe\(_2\)As\(_2\) (Al42622) \( (T_c = 27 \) K) \cite{14}. The inset of Fig. 2a) presents a systematic \( T \) evolution in \( ^{75}1/T_1 \) \( ^{75}T \) for a series of \( Y_x\)La\(_{1-x}\)1111 with \( x = 0.95, 0.2 \), and 0. In general, \( 1/T_1 \) is described as \( 1/T_1 \propto \sum_q |A_q|^2 \chi''(q, \omega) / \omega_0 \), where \( A_q \) is a wave-vector \( (q) \) dependent hyperfine-coupling constant and \( \chi(q, \omega) \) a dynamical spin susceptibility. Note that \( 1/T_1 \) is dominated by the low-energy limit of \( \chi''(q, \omega) / \omega_0 \) since an NMR frequency \( (\omega_0) \) is as low as a radio frequency. In \( Y_x\)La\(_{1-x}\)1111 compounds, a doping level of electrons into FeAs layers is expected to be similar, because an oxygen deficiency is nearly equivalent \cite{18}. In most electron-doped Fe-based SCs without magnetic rare-earth ions, Knight shift that is proportional to \( \chi(q, \omega) \) exhibits the slight decrease upon cooling \cite{18, 21, 24}. Hence, the increase of \( 1/T_1 \) can be attributed to the development of low-lying AFSFs with finite \( q \) below 150 K, which is more significant in going from \( x = 0 \) to 0.95 in \( Y_x\)La\(_{1-x}\)1111, as displayed in the inset. It should be noted that as the enhancement of low-lying AFSFs becomes visible upon cooling, \( T_c \) increases from 28 K to 34 K and then to 50 K at \( x = 0.0, 0.2, 0.95 \) in \( Y_x\)La\(_{1-x}\)1111, respectively. As shown in the figure, however, this trend is not always valid when looking at the data for Al42622 \( (T_c = 27 \) K) \cite{14} and BaK122 \( (T_c = 38 \) K) \cite{19}. Namely, \( 1/T_1 \) for both compounds markedly develop upon lowering tem-
perature in association with the stronger enhancement of AFSFs than that in Y0.95La0.051111, whereas Tc goes down to 38 K and 27 K for BaK122 and Al42622, respectively. Therefore, we remark that development of low-lying AFSFs due to the FS nesting is not always a unique factor for enhancing Tc, although an intimate correlation between the development of AFSFs and the enhancement of Tc is experimentally suggested in Fe-pnictides compounds such as Ba(Fe1-xCo_x)2As2(Ba122(Co)) [23], BaFe2(As1-xP_x)2(Ba122(AsP)) [28], FeSe [24, 26], and F-doped La1111 [27].

In order to shed light on SC characteristics, T1/1c normalized at Tc is plotted against the normalized temperature T/Tc in Fig. 2(b). The T1(Tc)/T1 for Y0.95La0.051111 decreases steeply as T < 0.4Tc, dominated by a large residual density of states (DOS) induced in nodal gaps under the large external field [26, 28]. The T1 result reveals that Y0.95La0.051111 is a nodeless high-Tc superconductor with Tc = 50 K.

Figure 2(a) presents the power n in the formula of 1/T1 ∼ T^n versus (Tc/T)1/1c/(RT)1/(RT)1/1c for various Fe-pnictide SCs. Here this formula is assumed in T range of 0.5Tc < T < Tc (see Fig. 2(b)). (T1)Tc and (T1)RT are the values at T = Tc and room temperature, respectively. (T1)Tc/(T1)RT > 1 means that AFSFs develop upon cooling, whereas (T1)Tc/(T1)RT < 1 points to the band structure effect near the Fermi level (see Fig. 2(a)). This remarkable correlation points to the fact that as AFSFs become more dominant in the normal state, the reduction rate in 1/T1 just below Tc, i.e. n, increases from n ∼ 3 for La1111 (Tc = 28 K) to n ∼ 7 for Al42622 (Tc = 27 K) [14], and hence the suppression of the coherence effect is more remarkable. These systematic evolutions of relaxation behaviors have allowed us to apply the multiple fully gapped s±-wave model based on the FS nesting to various Fe-pnictides. In previous works, we have presented a simulation of the relaxation behavior below Tc with a parameter αc of the coefficient of the coherence factor within the framework of the multiple fully gapped s±-wave model [14, 20]. In this simulation, αc = 1 is expected for sign-conserving intraband scattering and αc = −1 for sign-nonconserving interband scattering. In multiband systems, this value varies in the range −1 ≤ αc ≤ 1, dependent on the weight of their contribution in the nuclear relaxation process. Actually, as shown by the solid lines in Fig. 2(b), the previous experiments on Al42622 [14], BaK122[19], and La1111(HOVD) [20], were reproduced by reasonable parameters with αc ∼ −0.86, ~0, and ∼ +0.33, respectively. Here, it is valid to assume αc ∼ 0 for BaK122 since other parameters of SC-gap sizes and DOS are consistent with those values deduced by ARPES[30].

The result of Y0.95La0.051111 was also reproduced by taking parameters as ∆1/kpTc = 6.9(∆s/∆l=0.35), NFS/(NFS + NFS+)=0.67, smearing factor η = 0.05∆l, and αc ∼ 0. Here, SC gaps are represented as ∆l(∆s) and DOSs as NFS/(NFS+)(FS+) for FS with larger(smaller) gaps.

Next, we deal with these SC and normal-state behaviors in terms of the evolution of the Fermi surface (FS) nesting property based on band calculations reported thus far. Figures 3(b-e) present the schematic illustration of FS topologies, which are theoretically derived on the basis of the five-orbital model for several typical compounds, such as (b) Al42622 [24, 26], (c) Ln1111 with Tc higher than 50 K [37], (d) La1111(OPT) [37], and (e) La1111(HOVD).
result, $T_c$ goes down and the coherence effect is not significantly depressed [See Fig. 3(c)24]. By contrast, when $h_{\text{FN}}=1.5$ Å in Al24622 is longer than in other Fe pnictides, the size of the hole FS around $\Gamma'$ is larger, while one of two-hole FSs at $\Gamma(0,0)$ disappears, as shown in Fig. 3(b)24. In this case, since the FS nesting condition is much better between hole FSs ($\Gamma$ and $\Gamma'$) and electron FSs ($M[0,\pm\pi]$ and $[\pm\pi,0]$), AFSFs develop dramatically in such a manner that antiferromagnetic order could seemingly set in around 20 K. Even though the FS nesting is optimized, $T_c=27$ K is much lower than the highest $T_c=55$ K in Fe pnictides in association with the decrease of the FS multiplicity for Al24622, as argued below33.

Most remarkably, an important ingredient is that Ln1111 exhibiting $T_c$ higher than 50 K is characterized by three hole FSs; two of them are at $\Gamma$ and another is at $\Gamma'$, and two electron FSs at $M$ in the unfolded FS regime, as presented in Fig. 3(c). When noting that $h_{\text{FN}}\sim1.44$ Å in Y0.95La0.05111137 is longer than $h_{\text{FN}}\sim1.35$ Å for La0.8Y0.211111($T_c=34$ K) and $h_{\text{FN}}\sim1.33$ Å in La1111(OPT) ($T_c=28$ K), the appearance of $\Gamma'$ at $E_F$ causes the FS nesting condition to be better for Y0.95La0.051111 than that for La1111(OPT), resulting in the enhancement of AFSFs for the former. In this context, as the FS nesting condition becomes better, AFSFs are visible, and hence $T_c$ increases from 28 K in Ln1111, to 34 K in La0.8Y0.211111 up to 50 K in Y0.95La0.051111. Usui et al. have pointed out that the large FS multiplicity in Ln1111 in addition to the presence of AFSFs is another crucial factor for enhancing $T_c$ based on the spin-fluctuation mediated SC mechanism when the FeAs4 tetrahedron is close to a regular one realized in Ln111133. In this context, the optimized electronic state for the occurrence of SC in Fe pnictides is realized for the regular FeAs4 tetrahedron where multiorbital fluctuations may play some role in the onset of SC35, since spin and orbital degrees of freedom can be intimately coupled with one another. However, it is unlikely that multiorbital fluctuations become dominant to mediate high-$T_c$ SC in Y0.95La0.051111 because they prefer an $s_{\pm}$ wave SC38. Furthermore, we remark that the overall $T$ dependence of 1/$T_1$ in both SC and normal states is qualitatively accounted for by the fully-gapped $s_\pm$-wave SC model based on the FS nesting.

In conclusion, the $^{75}$As-NMR 1/$T_1$ measurement has revealed that Y0.95La0.051111 is the nodeless high-$T_c$ superconductor with $T_c=50$ K taking place under the moderately enhanced AFSFs due to the FS nesting condition better than in Ln1111 with $T_c=28$ K. We have demonstrated that these results are accounted for by the fully-gapped $s_\pm$-wave model based on the FS nesting33. We have proposed that the reason that $T_c\sim50$ K in Y0.95La0.051111 is larger than $T_c=28$ K in Ln1111 is that the FS multiplicity is maximized with the regular FeAs tetrahedron structure, and hence the FS nesting condition is better than that of La1111, developing moderately AFSFs. In the future, to address a mechanism of high-$T_c$ SC in Fe pnictides, it is desired to elucidate the $q$ and $\omega$ dependencies of $\chi''(q,\omega)$ under the spin and orbital degrees of freedom coupled with one another for Ln1111 with $T_c$ higher than 50 K.

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