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^{75}As NMR Study of the Oriented Pnictide Superconducting Compound $NdFeAsO_{0.83}F_{0.17}$

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Abstract. Magnetization and ^{75}As nuclear magnetic resonance (NMR) measurements in the superconductor $NdFeAsO_{0.83}F_{0.17}$ (T_C~46 K) are performed in order to investigate the effect of the Nd 4*f* electrons in the superconducting property. The magnetization curve displays a Nd 4*f* moments generated Curie-Weiss signal in the field of 7 T. ^{75}As NMR spectra in the oriented sample ($H_0 \| ab$) are recorded at 7 T in the temperature range 10–300 K and temperature dependent ^{75}As NMR shift, K_{ab} has been obtained. The K_{ab} curve shows a Curie-Weiss type contribution. The magnitude of hyperfine field, H_{hf} (4.4 kOe/μ_B) estimated from the K_{ab} vs χ plot indicates a non-negligible RKKY-type interaction between localized Nd 4*f* moments mediated by itinerant Fe 3*d* electrons.

Keywords: Iron-pnictides, NMR shift
PACS: 74.70.-b, 76.60.-k, 76.60.Es

INTRODUCTION

The family of oxypnictide superconductors with the general formula $REFeAsO_{1-x}F_x$ (RE=La, Ce, Pr, Nd, Sm, Gd, Tb, Dy) have generated a considerable attention in the recent years because they show high superconducting transition temperatures (T_C) and several similarities to the cuprates[1]. $LaFeAsO_{1-x}F_x$ is the first compound to show T_C~26 K for x=0.11[2]. Later, substitution of La^{3+} by other rare-earth (RE) ions resulted into higher T_C values[1]. A large scale effort on understanding the crystallographic, magnetic and superconducting properties has revealed that the FeAs layers contain all the electronic states near the Fermi energy. The superconducting response is attributed to the Fe 3*d* electronic state. X-ray and neutron scattering results have indicated a gradual shortening of the *c* axis when La^{3+} is replaced by other RE-ions and this observation has been corroborated with enhanced T_C. However, the role of the unpaired RE 4*f* electrons and their interaction with the Fe 3*d* electrons has not been understood explicitly. NMR, being a powerful microscopic tool, will be useful to address this issue. Earlier, the influence of the 4*f* electron on the magnetic behavior of the polycrystalline compounds of $CeFeAsO_{1-x}F_x$, $NdFeAsO_{1-x}F_x$ and $SmFeAsO_{1-x}F_x$ have been studied by Rybicki *et al*, [3] Jeglic *et al,*[4] and Prando *et al,*[5] respectively applying ^{75}As and ^{19}F NMR. Further elucidation of the magnetic properties of such superconductors demands measurements in single crystals. However, the synthesis of single crystals of pnictide superconductors is not frequently done. An alternative to a single crystals frequently applied for NMR measurements is to orient a large number of small crystallites by magnetic alignment method. Such samples actually have some advantages over a small single crystals since a large number of particles provides a strong signal. In the following, the study of the magnetic and superconducting property of $NdFeAsO_{0.83}F_{0.17}$ superconductor involving 4*f* electron is discussed in the light of ^{75}As NMR spectroscopy and magnetization measurements.

EXPERIMENATL DETAILS

Polycrystalline sample with nominal composition $NdFeAsO_{0.8}F_{0.2}$ was synthesized by two-step solid state reaction method using high-purity precursors of NdAs, FeF_2, Fe, and Fe_2O_3 as described in the reference 6. Powder X-ray diffraction of the sample confirmed the expected phases. Exact F-content of the compound has been measured by means of EMPA which showed the F-content of 17% instead of 20 %. The dc magnetic susceptibility, $\chi(T)$ was measured with a SQUID magnetometer in the presence of external magnetic fields of 5 Oe and 70 kOe and in the temperature range of 2–300 K in the heating cycle. To

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align the powder, the fine powder was mixed with an epoxy (Epotek 301) in the sample tube and then the sample tube kept fixed inside the NMR probe coil for a day inside the 7 T superconducting magnet. The NMR measurements of the grain aligned compound were carried out in a Bruker MSL100 pulse spectrometer at 7.0 T by a $\pi/2-\tau-\pi/2$ solid echo sequence.

RESULTS AND DISCUSSION

A. Magnetic Susceptibility

Temperature dependence of the zero-field-cooled (ZFC) $\chi(T)$ curves of $NdFeAsO_{0.83}F_{0.17}$ powder measured in 5 Oe and 70 kOe applied magnetic fields are shown in Fig. 1 (a, b). Fig. 1(a) shows the superconducting transition at T_C~46 K. Whereas $\chi(T)$ in Fig. 1(b) shows a substantial Curie-Weiss contribution.

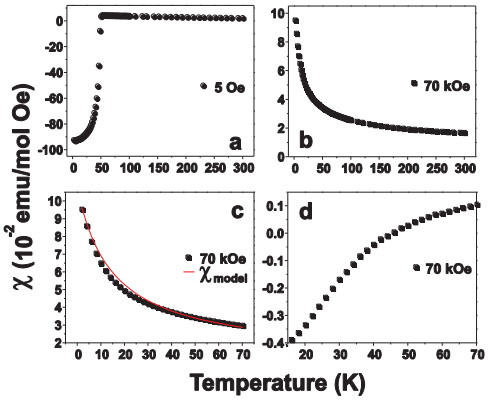

FIGURE 1. (a, b): Temperature dependence of ZFC $\chi(T)$ of $NdFeAsO_{0.83}F_{0.17}$ superconductor measured in 5 Oe and 70 kOe magnetic field, respectively. (c): Zoomed $\chi(T)$ curve in 70 kOe along with the fit to $\chi_{model}= \chi(0)+ \chi(Nd^{3+})$, traced by red solid line. (d): Difference $\chi(S)$ curve, zoomed around T_C, which is obtained by subtracting $\chi(0)+ \chi(Nd^{3+})$ contribution from the total experimental $\chi(T)$.

The $\chi(T)$ curve measured at 70 kOe has been fitted with $\chi_{model}=\chi(0)+\chi(Nd^{3+})$ as shown in the Fig. 1(c). Here, $\chi(0)$ is the temperature independent magnetic susceptibility and $\chi(Nd^{3+})=C/(T-\theta)$ is the Curie-Weiss susceptibility originated due the Nd^{3+} 4*f* moments. The best fit shown in the Fig. 1(c) gives $\chi(0)=0.012$ emu/mol Oe, the Curie constant $C=1.43$ emu K/mol and $\theta=-15\pm1$ K. The $C=1.43$ emu K/mol corresponds to an effective magnetic moment $\mu_{eff}=3.3\ \mu_B$ which is expected for localized Nd 4*f* moments. While $\theta=-15\pm1$ K resembles an antiferromagnetic interactions between the Nd 4*f* moments. In order to extract the superconducting response which is masked by the Curie-Weiss signal, we have subtracted $\chi(0)+\chi(Nd^{3+})$ from the total experimental susceptibility. This gives us $\chi(S)=\chi(T)-\chi_{model}$ and the temperature variation of $\chi(S)$ is shown in the Fig. 1(d).

B. ^{75}As NMR Spectra

Fig. 2(a) shows a frequency swept ^{75}As NMR central transition (-1/2↔1/2) line in the oriented sample of $NdFeAsO_{0.83}F_{0.17}$ recorded at 60 K. The measured NMR frequency of the central line for a typical quadrupolar powder spectra considering second order quadrupolar interaction and axial symmetry of the electric field gradient ($\eta=0$) can be written as,

$$\nu = \gamma H_0(1+K) + \Delta\nu_Q(\theta, H_0). \cdots\cdots (1)$$

Here γ is nuclear gyromagnetic ratio, K is the NMR frequency shift. Second term in the equation (1) is the second order quadrupolar shift and it results into a broad powder spectra with two horn peaks with comparable heights in the central transition.

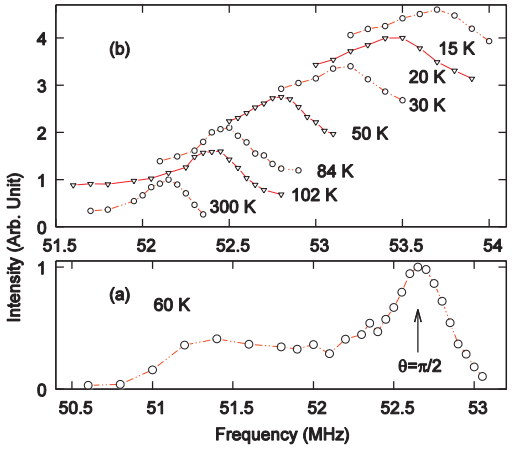

FIGURE 2. (a): Frequency swept ^{75}As NMR central transition in the oriented sample of $NdFeAsO_{0.83}F_{0.17}$ at 60 K. (b): Frequency swept ^{75}As NMR line corresponding to $\theta=90^\circ$ at different temperatures.

In $NdFeAsO_{0.83}F_{0.17}$, the peak appeared at high frequency side corresponds to $\theta=90^\circ$ with respect to external magnetic field direction (H_0). It is found that around 60% of the grains are aligned in the direction of $\theta=90^\circ$ which corresponds to $H_0 \perp c$ ($H_0 \| ab$). Since the alignment process worked reasonably well, the line width of the NMR signals in the oriented sample is reduced and it allowed us to record well resolved ^{75}As peaks in $NdFeAsO_{0.83}F_{0.17}$ at least down to 10 K as shown in the Fig. 2(b). It also shows that the peak position of the ^{75}As NMR line are gradually shifted to higher frequency with decreasing temperature. Moreover, gradual enhancements of NMR line widths has been observed.

C. ^{75}As NMR Shift

The resonance frequency ν in the oriented powder is given by $\nu=\nu_0+\nu_s+\nu_{quad}=\nu_0(1+K_{ab})+3\nu_Q^2/16\nu_0$ for $\eta=0$. The ν_{quad} in $REFeAsO_{1-x}F_x$ are generally considered as temperature independent. Hence the temperature dependency to the NMR line shift is entirely due to the magnetic shift K_{ab}. While K_{ab} estimation ν_{quad} amount has been deducted from the total NMR shift. Fig. 3(a, b) shows the temperature variation of K_{ab} which can be well described by the Curie-Weiss type behavior, $K(T)=K_0+K'C/(T-\theta)$ with K_0=-0.05%, Curie-Weiss temperature θ=-14 K, and K'=0.72% considering C=1.43 emu K/mol. The value of K' can be associated to the hyperfine interaction between ^{75}As nuclei and Nd 4*f* moments.

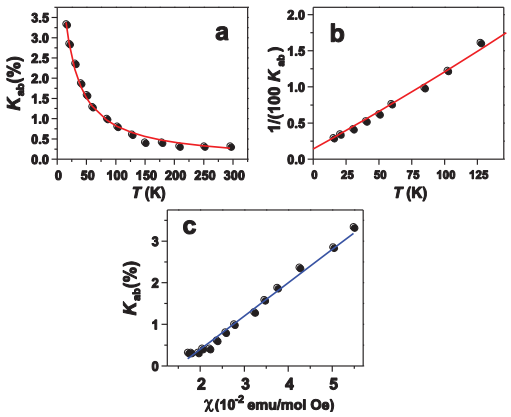

FIGURE 3. (a, b): Temperature variation of ^{75}As NMR shift, K_{ab} and $1/(100K_{ab})$ in the oriented sample of $NdFeAsO_{0.83}F_{0.17}$. The red solid lines represent a fit to Curie-Weiss model. (c): Variation of K_{ab} against $\chi(T)$. Solid line represents a linear fit with a slope of 0.8.

In fact, the ^{75}As NMR shift in the $NdFeAsO_{0.83}F_{0.17}$ is above two order larger than in $LaFeAsO_{1-x}F_x$ where the NMR shift is reflected only from the intrinsic contribution of Fe 3*d* electrons in the FeAs layer. The effect of the *f* electrons in addition with Fe 3*d* electrons increases NMR shift by one order larger $CeFeAsO_{1-x}F_x$ and $PrFeAsO_{1-x}F_x$ in comparison with $LaFeAsO_{1-x}F_x$.[1] ^{19}F NMR shift in $SmFeAsO_{0.85}F_{0.15}$ also showed a similar behavior. All these observations indicate an indirect RKKY-type exchange coupling among the Nd^{3+} magnetic moments in $NdFeAsO_{0.83}F_{0.17}$. To estimate the strength of the indirect exchange interaction we have plotted K_{ab} vs χ curve for 7 T. Fig. 3(c) shows the linear variation of K_{ab} with $\chi(T)$. The temperature dependence of the NMR shift K is directly related to the static uniform susceptibility χ by the following relation

$$k(T)=\frac{H_{hf}}{N_A\mu_B}\chi(T)+\delta.\cdots\cdots(2)$$

Where, H_{hf} is the hyperfine field experienced by the ^{75}As nuclei due to the localized Nd 4*f* electrons, which dominates the response function in the present case and δ is the chemical shift. Hence, a linear fit to the curve K_{ab} vs χ, leaving T as an implicit parameter, estimates H_{hf}=4.4 kOe/μ_B. The value of K'=0.72% leads to the value of hyperfine field, H_{hf}=4.0 kOe/μ_B which is close to the value H_{hf} obtained from K_{ab} vs χ curve. However, the value is much smaller than in $LaFeAsO_{0.9}F_{0.1}$[7] where H_{hf} arises due to coupling of ^{75}As nuclei with itinerant Fe 3*d* electrons.

SUMMARY

The magnetization curve in the $NdFeAsO_{0.83}F_{0.17}$ for H_0=5 Oe displays a superconducting transition at T_C~46 K and the superconducting response is suppressed at H_0=70 kOe by Nd 4*f* Curie-Weiss contribution. Magnetic alignment process fairly improved the NMR signal resolution and allowed us to record ^{75}As NMR spectra till 10 K. The magnitude of hyperfine field of H_{hf}=4.4 kOe/μ_B at ^{75}As nuclei suggests a weak RKKY-type hyperfine interaction between the *f* and conduction electrons. However, the conduction electrons which are interacting with *f* electrons should not be or weakly be involved in the pairing mechanism. This reconfirms previous conclusion that enhancement of T_C in Nd superconductors is associated with a size effect and not to a direct involvement of *f* electrons in the pairing mechanism.

REFERENCES

1. K. Ishida, Y. Nakai and H. Hosono, J. Phys. Soc. Jpn. **78**, 062001-062020 (2009).
2. Y. Kamihara, T. Watanabe, M. Hirano and H. Hosono, J. Am. Chem. Soc. **130**, 3296-3297 (2008).
3. D. Rybicki, T. Meissner, G. V. M. Williams, S. Chong, M. Lux, and J. Haase, J. Phys.: Condens. Matter **25**, 315701-315709 (2013).
4. P. Jeglic, J. –W. G. Bos, A. Zorko, M. Brunelli, K. Koch, H. Rosner, S. Margadonna and D. Arcon, Phys. Rev. B **79**, 094515-094521(2009).
5. G. Prando, P. Carretta, A. Rigamonti, S. Sanna, A. Palenzona, M. Putti, and M. Tropeano, Phys. Rev. B **81**, 100508-100511 (2010).
6. L. Malavasi, G. A. Artioli, C. Ritter, M. C. Mozzati, B. Maroni, B. Pahari, and A. Caneschi, J. Am. Chem. Soc., **132**, 2417-2420 (2010).
7. H. –J. Grafe, D. Paar, G. Lang, N. J. Curro, G. Behr, J. Werner, J. Hamann-Borrero, C. Hess, N. Leps, R. Klingeler, and B. Buchner, Phys. Rev. Lett., **101**, 047003-047006 (2008).