Muon spin relaxation studies of magnetic order and superfluid density in antiferromagnetic NdOFeAs, BaFe$_2$As$_2$ and superconducting (Ba,K)Fe$_2$As$_2$

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Zero-field (ZF) muon spin relaxation ($\mu$SR) measurements have revealed static commensurate magnetic order of Fe moments in NdOFeAs below $T_N \sim 135$ K, with the ordered moment size nearly equal to that in LaOFeAs, and confirmed similar behavior in BaFe$_2$As$_2$. In single crystals of superconducting (Ba$_{0.55}$K$_{0.45}$)Fe$_2$As$_2$, $\mu$SR spectra indicate static magnetism with incommensurate or short-ranged spin structure in $\sim 70\%$ of volume below $T_N \sim 80$ K, coexisting with a superfluid volume which exhibits superfluid-response consistent with nodeless gap below $T_c \sim 30$ K.

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The discovery of the iron oxypnictide superconductor La(O,F)FeAs ($T_c \sim 26$ K) [1] has triggered an unprecedented burst of quality research. Notable progress includes syntheses of materials with higher $T_c$'s [2, 3, 4] containing different Rare Earth (RE) elements, hole doping [5], reduced oxygen [6], and superconducting bi-layer AFe$_2$As$_2$ (A=Ba,Sr,Ca) systems by (A,K) substitution [7, 8] or application of pressure [9]. In characterization, one finds discoveries of commensurate antiferromagnetism of parent compounds LaOFeAs [10] and BaFe$_2$As$_2$ [11] by neutron scattering, nearly linear scaling of superfluid density and $T_c$ by muon spin relaxation ($\mu$SR) [12, 13, 14, 15], and studies of the electronic phase diagram by neutrons [16] and $\mu$SR [17], all of which exhibit remarkable similarities with the cases of cuprate systems. Very recent success in the fabrication of superconducting (Ba,K)Fe$_2$As$_2$ and (Sr,K)Fe$_2$As$_2$ single crystals [18, 19] has enabled detailed studies by ARCES [20, 21], and STM [22]. Quantum oscillation [23] has been observed in undoped crystals of BaFe$_2$As$_2$.

Several controversial issues, however, remain open to further studies. Neutron measurements found antiferromagnetic order of Fe moments below $T \sim 135$ K in LaOFeAs [10] and CeOFeAs [16] with the ordered Fe moment size of 0.36 and 0.8 Bohr magnetons, respectively. Bos et al. and Qiu et al. [24], however, reported absence of corresponding antiferromagnetic order in NdOFeAs. Such a drastic dependence on RE elements is surprising, and has to be re-examined by other magnetic probes. Reports on pairing symmetry are divided between those favoring a gap with [22, 23] or without [21, 20] nodes.

Extensive characterization of single crystal specimens is very important in the early stage of materials development for production of crystals with improved qualities in the future. To address these issues, we have performed $\mu$SR measurements on ceramic specimens of NdOFeAs and BaFe$_2$As$_2$, as well as on superconducting single crystals of (Ba$_{0.55}$K$_{0.45}$)Fe$_2$As$_2$. As reported in this letter, our results demonstrate antiferromagnetism of the former two systems nearly identical to that of LaOFeAs. In the superconducting single crystals, we found coexisting signals due to incommensurate or short-ranged static magnetism from a partial volume fraction, and to a superfluid response from the remaining volume.

Ceramic specimens of NdOFeAs and BaFe$_2$As$_2$, with dimensions of 8-10 mm in diameter and 1-2 mm thick, were synthesized at IOP in Beijing following the methods published elsewhere [8, 27]. Single crystals of (Ba$_{0.55}$K$_{0.45}$)Fe$_2$As$_2$ were prepared at Ames Lab., Iowa following the method described in ref. [18]. Small crystallites, with a typical size of $2 \times 2 \times 0.1$ mm, were aligned with their main surface (ab-plane) parallel to each other to form a mosaic specimen of 65 mg in weight covering the area of $\sim 7$ mm in diameter. $\mu$SR measurements were performed at TRIUMF, Vancouver, with the beam direction perpendicular to the main surface of each specimen, following a standard method described in refs. [30, 31, 32].

In zero-field (ZF) $\mu$SR measurements of NdOFeAs, long-lived and single-component muon spin precession was observed below $T_N \sim 135$ K, as shown in Fig. 1(a). The precession amplitude corresponds to the value expected for $\sim 70$-80 % of muons. The temperature de-
dependence of the frequency is shown in Fig. 1(b). The anomaly of the frequency below $T = 5$ K is presumably due to magnetic ordering of Nd moments, which was detected by neutron scattering [24]. The frequency in NdOFeAs is nearly equal to that observed in LaOFeAs [14] at $T > 5$ K, indicating that the ordered Fe moment for these two systems are the same size for temperature regions unaffected by the Nd ordering. Figure 1(c) shows the fraction of muons in a paramagnetic environment derived from the precession asymmetry observed in weak transverse field (WTF) of 100 G and also from the ZF-$\mu$SR spectra. This figure indicates that a small fraction ($\sim 20-30\%$) of muons remain in paramagnetic environment even below $T_N$, which could either be due to a minor paramagnetic volume fraction or to possible cancellation of the static local field at corresponding muon sites from symmetry reasons. Despite this uncertainty, the overall results in Fig. 1 clearly demonstrate commensurate antiferromagnetic order in most of the volume fraction of NdFeAsO below $T_N$.

Figure 2(a) shows ZF-$\mu$SR spectra in BaFe$_2$As$_2$. Below $T_N = 140$ K, we find long-lived oscillations with two frequencies: the main frequency at 28.8 MHz from 80 % of the muons and the other at 7 MHz from 20 %, at $T \rightarrow 0$ as shown in Fig. 3(a). The total volume fraction of magnetically ordered region is essentially 100 % below $T_N$ (Fig. 3(b)). Static magnetic order of this system was first identified by Rotter et al. [33] who observed a Moessbauer hyperfine field $H_{\text{Moess}}$ of 5.47 T at $T \rightarrow 0$ which corresponds to an ordered Fe moment of 0.4 Bohr magnetons. In the mono-layer parent system LaOAsFe, the Moessbauer field $H_{\text{Moess}}$ was 4.86 T [34], while the main frequency in $\mu$SR was 23.0 MHz at $T \rightarrow 0$ [14, 34]. The ratio of main $\mu$SR frequencies $28.9/23.0 = 1.25$ of the bi- and mono-layer systems is nearly equal to the ratio $5.47/4.86 = 1.13$ of $H_{\text{Moess}}$, indicating that the effective hyperfine coupling constants for muons in these two systems are close to each other within 15 %. Combined $\mu$SR, Moessbauer and neutron results demonstrate...
that antiferromagnetism of BaFe$_2$As$_2$ is nearly identical to that of LaOFeAs and NdOFeAs in terms of $T_N$, spin structure $^{10,11}$, ordered moment size, and (nearly full) ordered volume fraction.

In the K-doped single crystals (Ba$_{0.55}$K$_{0.45}$)Fe$_2$As$_2$, we observed ZF-$\mu$SR spectra with a damped precession signal below $T = 70 - 80$ K, as shown in Fig. 2(b), only when the initial muon polarization is perpendicular to the c-axis direction. This implies an onset of highly inhomogeneous static field, parallel to the c-axis, at the muon site(s). Although the oscillating spectra can be fit either to a damped cosine or damped Bessel function, the Bessel-fit (solid line in the inset of Fig. 2(b)) gives the initial phase $\phi \approx 7$ degree at time $t = 0$ consistent with the experimental condition, unlike the cosine-fit with $\phi \approx -35$ degrees which is significantly off-phase. This suggests a possibility of incommensurate and/or stripe spin structure of the ordered region $^{32,33,36}$. Note that a similar damped Bessel signal was observed also in 3% F doped La(O$_{0.07}$F$_{0.03}$)FeAs $^{14}$.

Figure 3(a) shows the precession frequency in the K doped crystals, which is close to that in undoped BaFe$_2$As$_2$. This indicates a gradual evolution of spin configuration with K doping. The fraction of muons in a paramagnetic environment in Fig. 3(b), derived from the corresponding asymmetry of the $\mu$SR spectra in WTF and ZF, demonstrates that the magnetic order in (Ba$_{0.55}$K$_{0.45}$)Fe$_2$As$_2$ is detected by $V_m$, $\sim 60-80%$ of the muons below $T \sim 60$ K, which implies the volume fraction $V_m \sim 50-70%$ of the region with static magnetism $^{32}$.

The remaining paramagnetic signal allowed measurements of the superfluid response below $T_c \sim 30$ K in (Ba$_{0.55}$K$_{0.45}$)Fe$_2$As$_2$ single crystals. The inset of Fig. 4 shows the muon spin relaxation rate $\sigma$ observed in transverse external fields of 100 and 500 G parallel to the c-axis, obtained after quadratically subtracting the background relaxation due to nuclear dipolar fields from the observed Gaussian relaxation rate $\sigma$ shown in the inset. The temperature dependence is compared to BCS theory for isotropic s-wave pairing (broken line) and scaled results from YBCO cuprate superconductor $^{30}$ (small open diamond symbols).

The observed $\sigma_{sc}(T \rightarrow 0) \sim 0.33$ $\mu$s$^{-1}$ for crystal specimens imply that the superfluid density in the present (Ba$_{0.55}$K$_{0.45}$)Fe$_2$As$_2$ specimens is about 30% of that in La(O,F)FeAs with comparable $T_c$. The small superfluid
density and a large volume with static magnetism suggest that the system is a mixture of regions with static magnetic order and remaining regions with superconductivity. This picture is consistent with the results of the specific-heat jump reported in ref. [18], as well as STM studies which found two distinct responses [22].

Structural and magnetic phase transition at $T = 85 K$ was found in single crystals of undoped BaFe$_2$As$_2$ prepared with the Sn-flux method [18]. The magnetic order below $T \approx 80 K$ in the present K-doped crystals, prepared with the same method, suggests a possibility of a wide spatial spread of K concentrations, even beyond $\pm 7\%$ layer-by-layer spread around 45 $\%$ nominal K concentration found in ref. [18]. At this moment, however, it is not clear whether the superconducting and magnetically ordered regions are patterned as a microscopic phase separation, similarly to oxygen-overdoped LSCO cuprates [32], or as a segregation of a more macroscopic length scale.

The present studies are made on the first set of superconducting single crystals ever produced among the bilayer iron-based systems. Therefore, the results should be received with a caution that improved quality / homogeneity / size of the specimen and improved statistics of data could possibly alter the essential message, as has been experienced for the cases of cuprate systems. Within such limitations, however, the present results indicate: (a) decent superfluid response of the crystals, (b) possibility of intrinsic microscopic coexistence of regions with and without static magnetic order, (c) nodeless energy gap, which might be intrinsic or be related to scattering of carriers due to magnetic volumes, and (d) possibility of involvement of incommensurate or stripe spin correlations near the border of antiferromagnetic and superconducting states.

Our $\mu$SR results on NdOFeAs are clearly inconsistent with the neutron studies of NdOFeAs [24] which found an absence of corresponding Fe ordering. Neutron measurements in BaFe$_2$As$_2$ [11] and LaOFeAs [10] estimate the size of the ordered Fe moment to be 0.87 and 0.36 Bohr magnetons, respectively. In contrast, $\mu$SR and Moessbauer studies found a comparable moment size in these two systems. The origin of these disagreements is unclear at this moment. For the estimate of ordered moment size, however, local probes like $\mu$SR and Moessbauer generally provide more accurate information than volume-integrated Bragg-peak intensity of neutron scattering.

In summary, our $\mu$SR measurements of Fe-based high-$T_c$ systems have revealed static magnetic order of NdOFeAs, demonstrated similar magnetic behavior in parent compounds of mono- and bi-layer systems NdOFeAs, LaOFeAs, and BaFe$_2$As$_2$, and elucidated coexisting static magnetism and superconducting responses in single crystals of (Ba$_{0.15}$K$_{0.45}$)Fe$_2$As$_2$.

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