Static Magnetic Order and Superfluid Density of RFeAs(O, F) (R=La, Nd, Ce) and LaFePO Studied by Muon Spin Relaxation: Unusual Similarities with the Behavior of Cuprate Superconductors

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Muon spin relaxation measurements in iron-oxypnictide systems have revealed: (1) commensurate long-range order in undoped LaFeAsO; (2) a Bessel function line shape in LaFeAs($O_{0.97}F_{0.03}$) which indicates possible incommensurate or stripe magnetism; (3) anomalous weak magnetism existing in superconducting LaFePO, CeFeAs($O_{0.084}F_{0.16}$), and NdFeAs($O_{0.88}F_{0.12}$) but absent in superconducting LaFeAs($O_{0.92}F_{0.08}$); and (4) scaling of the superfluid density with T_c in the Ce-, La-, and Nd-FeAs superconductors following a nearly linear relationship found in cuprates.

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A renewed interest in high-T_c superconductivity has been generated by the recent discoveries of iron-oxypnictide superconductors LaFePO ($T_c \sim 5$ K) [1] and LaFeAs(O,F) ($T_c \sim 26$ K) [2], followed by the subsequent development of materials with higher T_c's up to ~55 K containing rare-earth-metal (R) elements, such as Ce, Nd, and Sm [3–5] instead of La. Carrier doping is achieved by (O,F) or (La,Sr) [6] substitutions as well as high-pressure oxygen synthesis [5]. For an overall understanding of the mechanisms of high-T_c superconductivity, it would be very instructive to compare these new superconductors with the cuprate systems. Muon spin relaxation (μSR) studies have provided unique information on magnetic order [7–9] and superfluid density [10–13] in cuprate systems. In this Letter, we report μSR measurements on the undoped and doped iron-oxypnictide systems LaFePO, LaFeAsO, LaFeAs($O_{0.97}F_{0.03}$), LaFeAs($O_{0.92}F_{0.08}$), CeFeAs($O_{0.84}F_{0.16}$), and NdFeAs($O_{0.88}F_{0.12}$). Our results demonstrate several generic features common to the iron oxypnictides and cuprate systems, including long-range commensurate antiferromagnetism of the undoped parent compounds, incommensurate or stripe magnetism of lightly doped systems near the border of magnetic and superconducting phases, and scaling of the superfluid density with T_c following a nearly common linear relationship.

Polycrystalline specimens of all of the FeAs compounds were synthesized at the Beijing National Laboratory for Condensed Matter Physics, following the method described in [14]. The LaFePO specimens were synthesized at Osaka University, in silica tubes using LaP, Fe, and Fe_2O_3 as starting materials. These specimens were made into ceramic pellets 8 mm in diameter and 1–2 mm thick. The specimens of undoped LaFeAsO and superconducting LaFeAs($O_{0.92}F_{0.08}$) are identical to those used in neutron scattering measurements [15] which revealed collinear antiferromagnetic order of LaFeAsO below ~134 K and absence of long-range magnetism in superconducting LaFeAs($O_{0.92}F_{0.08}$). μSR measurements were performed at TRIUMF using a He gas-flow cryostat in zero field (ZF), a longitudinal field (LF), and a weak transverse field (WTF) to characterize magnetism and in a transverse field (TF) to determine the magnetic field penetration depth. Details of the μSR method can be found in Refs. [12,13].

Figure 1(a) shows the time spectra of the muon spin polarization in zero field observed in the undoped parent system LaFeAsO. The long-lived and very clear muon spin precession signal indicates a spatially long-ranged and homogenous magnetism, consistent with the commensurate Bragg peak for the collinear antiferromagnetic structure found by neutron studies [15]. The μSR spectra can be fit to the sum of a high (low) frequency $f \sim 23$ MHz (3 MHz) signal at $T \rightarrow 0$ with a dominant ~60% (minor ~10%) signal amplitude, and the remaining $1/3$ component which exhibits a slow $1/T_1$ relaxation. The temperature dependences of these frequencies are shown in Fig. 2(a). These results in LaFeAsO are essentially consistent with earlier reports by μSR and Moessbauer studies [16,17] in LaFeAsO.

ZF μSR spectra of the 3% doped LaFeAs($O_{0.97}F_{0.03}$), shown in Fig. 1(b), exhibit much faster damping than those in undoped LaFeAsO. The fast-damping spectra in Fig. 1(b) fit well to Bessel functions (multiplied by a

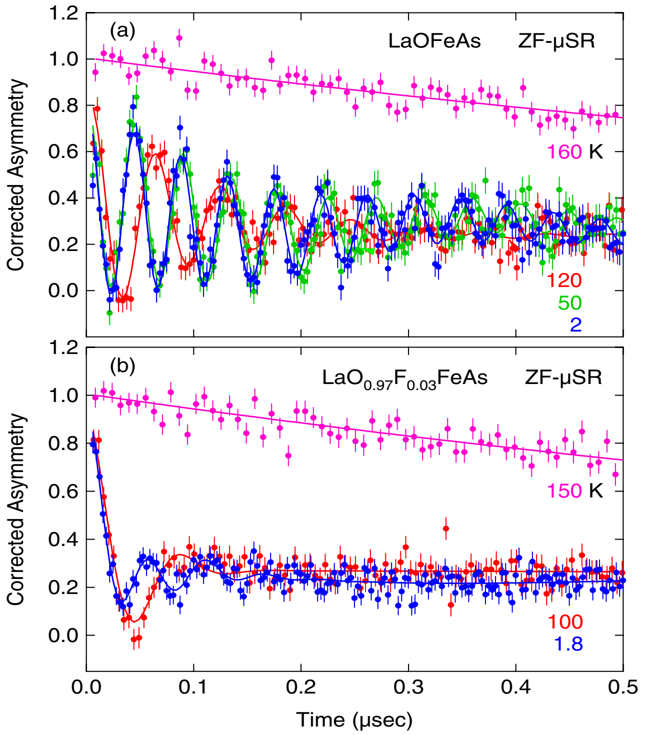

FIG. 1 (color online). Time spectra of zero-field μSR in (a) undoped LaFeAsO and (b) 3%-doped LaFeAs($O_{0.97}F_{0.03}$). The solid line in (b) for $T = 100$ K represents a single Bessel function multiplied by a Gaussian damping, while the $T = 2$ K data were fit to the sum of three Bessel*Gaussian signals.

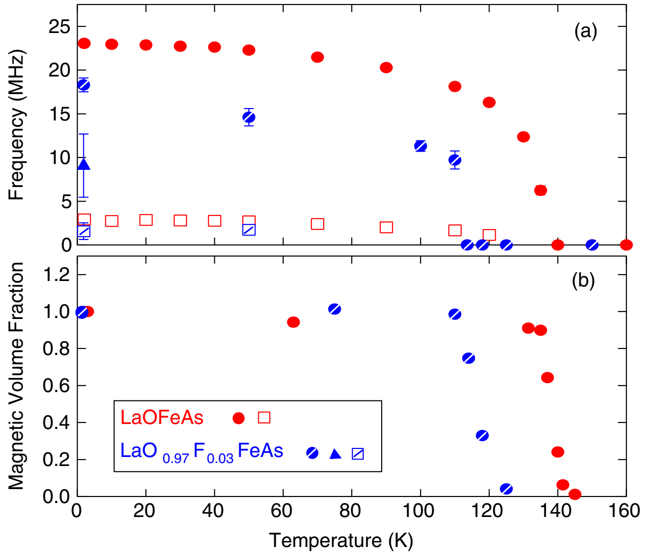

FIG. 2 (color online). (a) Muon spin precession frequency observed in zero field in LaFeAsO (for two cosine signals) and LaFeAs($O_{0.97}F_{0.03}$) (for one to three Bessel signals). (b) The magnetic volume fraction estimated by WTF measurements with the external field of 50 G.

Gaussian damping factor), which are shown by the solid lines. The Bessel function line shape was first observed in ZF μSR in an incommensurate spin density wave (SDW) system $(TMTSF)_2PF_6$ [18] and subsequently in $La_{1.875}Ba_{0.125}CuO_4$ [19], $La_{1.47}Nd_{0.4}Sr_{0.13}CuO_4$, and several other cuprate systems [9] which undergo formation of static spin stripes. The comparison of parent and doped systems in Fig. 1 exhibits exactly the same features as in the case of the cuprates, La_2CuO_4, and the 1/8 doped spin stripe systems, shown in Figs. 4(b), 2(a), and 2(b), respectively, of Ref. [9]. On the other hand, it is also possible to expect a highly damped spectrum in a commensurate antiferromagnet with substantial randomness. Neutron scattering studies are required for a clear distinction of these three spin structures and determination of the spatial periodicity.

Figure 2(a) compares the ZF precession frequencies of the two cosine signals in undoped LaFeAsO (closed and open red symbols) with the frequencies of the Bessel function signals in the 3%-doped compound (closed blue symbol). In the case of incommensurate SDW order, the frequency of the Bessel function corresponds to the internal field at the muon site near the maximum SDW amplitude. The lower Bessel frequencies at $T = 2$ K in the 3%-doped system implies that the average internal field at the muon site is substantially reduced from that in the undoped LaFeAsO. This is reminiscent of the case of cuprates, where the ZF μSR frequency in La_2CuO_4 was about ~30% higher than the Bessel frequency in the 1/8 doped stripe systems, as shown in Fig. 3(b) of Ref. [9]. Such a large variation of internal field via 3% carrier doping is hardly expected by a simple magnetic dilution of commensurate antiferromagnets. These considerations suggest a possibility of incommensurate or static stripe magnetism in LaFeAs($O_{0.97}F_{0.03}$).

Figure 2(b) shows the volume fraction of the magnetically ordered region in the undoped and doped FeAs systems, obtained in WTF μSR measurements [20] with WTF ~50 G, after a correction for a background signal with a spectral weight of less than 10% of the total signal amplitude. In both doped and undoped systems, magnetic order develops in essentially the full volume fraction below the Néel temperature T_N. A continuous change in the finite volume fraction was observed in a narrow temperature range within ±5 K of T_N in both systems, indicating that the phase transitions are weakly first-order in both cases. A rather high $T_N \sim 110$ K and full magnetic volume fraction of the doped system indicate robustness of static magnetism against carrier doping. This is reminiscent of the case of nonsuperconducting electron-doped cuprates.

ZF μSR measurements have also revealed an anomalous and weak relaxation existing in the normal and superconducting states of LaFePO, CeFeAs($O_{0.84}F_{0.16}$), and NdFeAs($O_{0.88}F_{0.12}$), as well as in the paramagnetic state of LaFeAsO and LaFeAs($O_{0.97}F_{0.03}$) above T_N. As shown by the time spectra in Figs. 3(a)–3(c), this relaxation is seen in most of the systems in the present study, except for the superconducting LaFeAs($O_{0.92}F_{0.08}$). The anomalous relaxation has also been reported in $SmO_{0.082}F_{0.18}FeAs$ [21], while the absence of the effect in the La-based super-

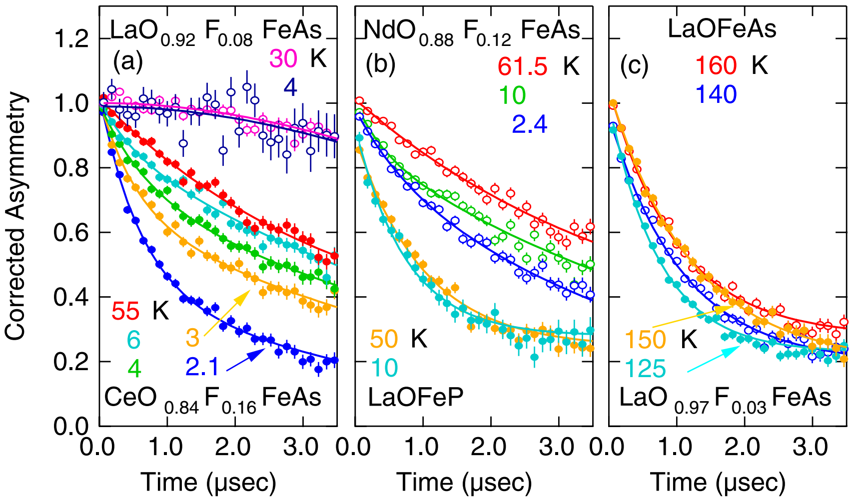

FIG. 3 (color online). Muon spin relaxation function observed in zero field in (a) La and Ce-FeAs superconductors, (b) Nd-FeAs and LaFePO superconductors, and (c) in the paramagnetic state of LaFeAsO and LaFeAs($O_{0.97}F_{0.03}$).

conducting system is consistent with an earlier report [22]. These ZF μSR spectra exhibit almost no temperature dependence, except for the Ce compound below $T \sim 4$ K and the Nd compound at $T = 2.4$ K, which is presumably due to imminent ordering of Ce or Nd moments.

Generally, muon spin relaxation in ZF can be due to (1) a random static field (nuclear dipolar field, etc.) which causes dephasing of muon spins and (2) dynamic fields from spin fluctuations or spin waves resulting in energy-dissipative $1/T_1$ processes. (1) and (2) can be distinguished by their different responses to LF [13]. The static origin of the anomalous relaxation was confirmed by LF μSR measurements in LaFePO at $T = 8$ and 2 K and in our unpublished work on nonsuperconducting CeFeAs($O_{0.14}F_{0.06}$), while static and additional dynamic effects were found for the Sm compound in Ref. [21] at $T = 60$ K. The observed relaxation rate of 0.3–1 μs^{-1} corresponds to a random static internal field of about 3–10 G (which is significantly larger than nuclear dipolar fields expected in these systems). If this field comes from dilute frozen moments of ~1 Bohr magneton, it corresponds to a 0.1%–0.5% concentration level per formula unit. These features indicate: (1) the slow anomalous relaxation does not necessarily correlate with the superconducting transition; (2) this effect exists not only in systems containing magnetic rare-earth elements but also in several La-based systems; (3) dilute frozen moments, conceivable for systems containing minor impurity phases or nonstoichiometric Fe and rare-earth concentrations, are possible origins of the observed effect. A magnetization study of LaFePO [23] found a signature of FeP impurity. The reported absence of superconductivity in stoichiometric LaFeAsO suggests that off-stoichiometry of O may be required for bulk superconductivity of this system.

On the four superconducting specimens [those in Figs. 3(a) and 3(b)], we performed TF μSR measurements in TF = 300, 600, and 1200 G to measure the magnetic field penetration depth λ and to derive the superfluid density n_s/m^* (superconducting carrier density/effective mass). The superfluid density is proportional to the relaxation rate σ, which reflects a dephasing process due to the inhomogeneous field distribution from the flux vortex lattice below T_c. To correct for the effect of the anomalous magnetic relaxation shown in Fig. 3, we first obtained an exponential relaxation rate in TF above T_c, fixed this value in the fitting, multiplied this exponential damping by a Gaussian precession envelope $\exp(-\sigma^2 t^2/2)$, and then derived σ as shown in Fig. 4. There is essentially no difference in this procedure and fitting to a simple Gaussian function in the FeAs-based superconductors, since the prefixed exponential relaxation rates (0.03, 0.24, and 0.34 μs^{-1}, respectively, for the La, Nd, and Ce compounds) are much smaller than the superconducting Gaussian relaxation rates. For LaFePO, the exponential (1.36 μs^{-1}) and Gaussian relaxation rates are very close, resulting in a significant systematic uncertainty in accuracy of the superfluid density. The rapid increase of σ in the Nd compound (at $T = 2$ K) and the Ce compound (below $T = 4$ K) are presumably due to imminent magnetic order of moments on these elements. Otherwise, $\sigma(T)$ shows a

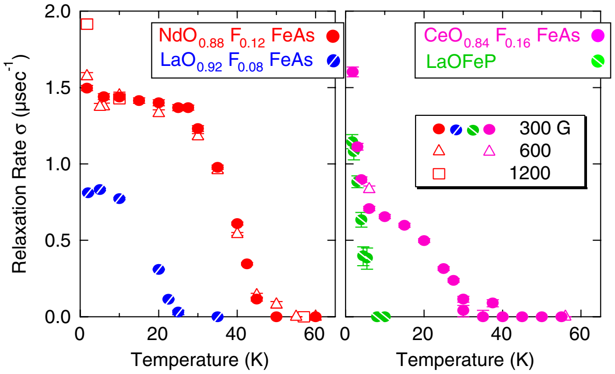

FIG. 4 (color online). Muon spin relaxation rate σ observed in transverse external fields in superconducting FeAs- and FeP-based systems. The relaxation envelope was fit to a prefixed exponential function multiplied by a Gaussian function $\exp(-\sigma^2 t^2/2)$.

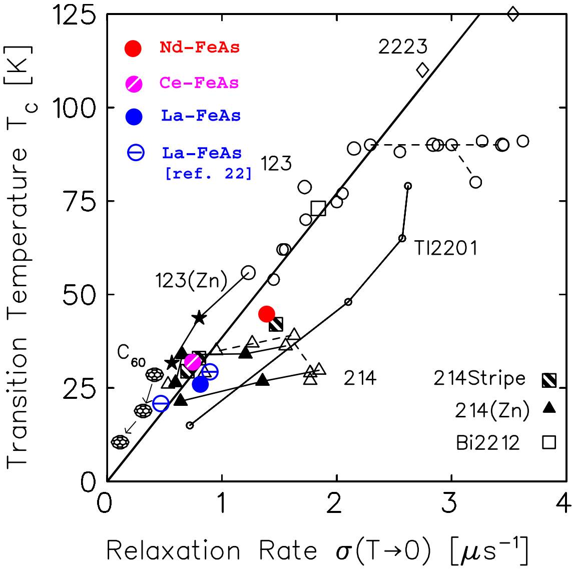

FIG. 5 (color online). A plot of the muon spin relaxation rate $\sigma(T \to 0) \propto n_s/m^*$ versus T_c for the iron-oxypnictide systems (colored solid circles for the present results and colored open circles for Ref. [22] compared with the results for the cuprates and alkali-doped C_{60} systems [10,25,26]). The FeAs superconductors follow the linear relationship found for underdoped cuprates in this plot.

saturation at low temperatures, consistent with an earlier report [22]. In view of the difficulty in determining the pairing symmetry using μSR results on ceramic specimens experienced in the cuprates, however, we postpone the symmetry arguments until results on single-crystal specimens become available.

In the $\sigma(T \to 0)$ vs T_c plot of Fig. 5, we compare the results of the FeAs-based systems to cuprates and a few other exotic superconductors. Three points from the present study and two other points from Ref. [22] demonstrate that the electron-doped FeAs systems follow a nearly linear relationship between T_c and $n_s/m^* \propto \sigma(T \to 0)$, with the slope approximately the same as that found for the cuprates. We note that our earlier results on an electron-doped cuprate $(Nd, Ce)_2CuO_4$ [24] also follow the behavior of hole-doped cuprates. Figure 5 demonstrates that many type-II superconductors, including cuprates, iron oxypnictides, and C_{60} systems, share approximately the same ratios between their superfluid energy scales and T_c. This implies that n_s/m^* is an important factor in determining T_c.

A large relaxation rate $\sigma(T \to 0)$ observed in the superconducting system LaFePO with a rather low T_c, however, suggests that an additional factor(s), such as close proximity to the magnetically ordered state, also plays a significant role in determining T_c, as discussed in Refs. [25,26] for cuprates. In these references, one of us proposed a pairing mechanism for cuprates based on a charge motion (with an effective Fermi energy kT_F derived from the superfluid density) resonating with antiferromagnetic spin fluctuations (with an energy scale $\hbar\omega_{AF}$ represented by the exchange interaction J) as $kT_F \sim \hbar\omega_{AF}$. With the present results suggesting $\sigma(T \to 0) \propto n_s/m^* \propto kT_F$ (in 2D systems) $\propto T_c$, this resonant spin-charge motion model might also be applicable to oxypnictides if their $\hbar\omega_{AF}$ is approximately 50 meV or so, as estimated in a theoretical work [27]. In this model, superconductivity should be absent in the doping region with static Cu/Fe magnetism (except for phase separation), which is the case in both the copper oxide and FeAs superconductors. In summary, we demonstrated several common features between cuprates and oxypnictides by comparing μSR results on their magnetism and superfluid density.

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