Anisotropic structural and magnetic properties of the field-aligned superconducting system
SmFeAsO\(_{1-x}\)F\(_x\) (\(x = 0, 0.1, 0.2, 0.25 \text{ and } 0.3\))

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Abstract. Anisotropic structural and magnetic properties of the field-aligned superconducting system SmFeAsO\(_{1-x}\)F\(_x\) (\(x = 0, 0.1, 0.2, 0.25 \text{ and } 0.3\)) are reported. Due to the Fe spin-orbital related anisotropic exchange coupling, all the tetragonal microcrystalline powders in epoxy were aligned at room temperature using the field-rotation method where the tetragonal \(ab\)-plane is parallel to the magnetic alignment field \(B_a\) of 0.9 T and the \(c\)-axis parallels to the rotating axis. Anisotropic magnetic properties are studied through low temperature magnetic measurements along the \(c\)-axis and paralleled to the \(ab\)-plane of aligned samples in both zero-field-cooled (ZFC) and field-cooled (FC) modes. The under-doped compound (\(x = 0.1\)) is not superconducting with an antiferromagnetic Néel temperature \(T_N \approx 40\) K, while the two optimum-doped compounds (\(x = 0.2\) and 0.25) show high superconducting transition temperatures \(T_c\) of 49K and 50K, respectively. The variation of anisotropic structural and magnetic properties for this system are discussed and compared with the previously reported 52 K anisotropic superconductor Sm\(_{0.95}\)La\(_{0.05}\)FeAsO\(_{0.85}\)F\(_{0.15}\).

1. Introduction
High-\(T_c\) superconducting iron-based \(R\)FeAsO\(_{1-x}\)F\(_x\) (rare earth \(R = \) La, Ce, Pr, Nd, or Sm) system with transition temperature up to 55 K were reported since 2008\(^1\)\(^3\)\(^6\). The ZrCuAsSi-type tetragonal structure (space group \(P4/nmm\)) is a layered structure where the metallic FeAs layer is separated by the insulating RO\(_{1-x}\)F\(_x\) layer. The parent compound LaFeAsO is a normal semi-metal which shows a spin density wave (SDW) type antiferromagnetic order below 150 K due to Fermi surface nesting with a tetragonal to orthorhombic structural transition\(^2\). Electron doping to the FeAs layer through \(F^-\)-substitution in the \(O^{2-}\) site or \(O^{2-}\)-deficiency suppresses both the magnetic order and the structural distortion in favor of superconductivity\(^1\)\(^3\). On the other hand, hole doping through \(Sr^{2+}\)-substitution in the \(R^{3+}\) site gives similar effect\(^4\). Therefore, similar to the high-\(T_c\) cuprate systems, the superconductivity in these iron-based compounds occurs in close proximity to a long range antiferromagnetic ground state.

Since the FeAs layer is the superconducting layer of the \(R\)FeAsO\(_{1-x}\)F\(_x\) system, studies on the anisotropic magnetic properties and their relation to the structural transition are crucial to understanding this new iron-based system. However, high-quality single crystal is difficult
to acquire, a much simpler way using the field-rotation alignment method presents a practical mean to study the anisotropic magnetic and structural transition properties[5].

In this report, SmFeAsO$_{1-x}$F$_x$ ($x = 0, 0.1, 0.2, 0.25, 0.3$) powders are field-aligned in epoxy at room temperature and magnetic susceptibility were measured along the $ab$-plane and $c$-axis in a 10-G external magnetic field. Lattice parameters are determined from x-ray diffraction data and systematic variations on lattice aspect ratio $c/a$ and cell volume $V = a^2c$ are presented.

2. Experimental

The polycrystalline SmLaFeAsO$_{1-x}$F$_x$ powder samples were prepared by the conventional solid state reaction method. First the SmAs powder was prepared by reacting Sm and As powders at 650°C for about 5 hours in an evacuated and sealed quartz tube, and then fine powders of SmAs, FeAs, Fe, LaF$_3$, Fe$_2$O$_3$ were mixed together according to the stoichiometric ratio of SmLaFeAsO$_{1-x}$F$_x$, ground thoroughly, and then pressed into pellets. The pellets were further wrapped in Ta foil to be sealed in an evacuated quartz tube, and annealed at 1,160°C for about 50 hours.

The SmLaFeAsO$_{1-x}$F$_x$ were further grounded into powders with average grain sizes ranged between 1 to 10 µm, and were mixed with epoxy in a quartz tube ($\phi = 8$ mm) with a powder-to-epoxy ratio of 1 to 5. The quartz tube filled with powder-in-epoxy was immediately placed in a 0.9-T magnetic field at room temperature and rotated at a speed of 10 rpm for 4 hours (time for epoxy curing) with the rotation axis perpendicular to the 0.9-T applied magnetic field such that the tetragonal $ab$-plane is parallel to the applied magnetic field and $c$-axis is parallel to the rotation axis[5]. The cured cylindrical compounds were sliced cross-wise into a round button to prepare a surface for x-ray diffraction study on the $ab$-plane, and then sliced perpendicularly to the surface button to prepare another surface for x-ray study along $c$-axis.

Magnetic susceptibility data were collected with a Quantum Design 1-T $\mu$-metal shielded MPMS$_2$ or a 7-T MPMS superconducting quantum interference device (SQUID) magnetometer from 2 K to 300 K.

3. Results and Discussions

The diffraction patterns of both random and aligned powders were compared to confirm proper orientations for further magnetic measurements. Enhanced $(hk0)$ diffraction lines ($h + k = 2n$) were expected and observed for surfaces parallelled to the $ab$-plane, and enhanced (001) diffraction lines, for surface parallelled to the $c$-axis. The lack of the forbidden line (210) is consistent with the space group $P4/nmm$. The relative intensity of the (102) line which may be due to intrinsic weak magnetic anisotropy, low aligned field (0.9 T), imperfect powder preparation or alignment procedure gives estimated degrees of alignment of 70-80% for $x = 0.25$ and 0.3 samples and of 80-90% for $x = 0.1$ and 0.2 samples.

For field-aligned compounds with F content $x = 0.2, 0.25$, and 0.3, molar magnetic susceptibility $\chi_{mol}$ of aligned powder in an applied magnetic field $B_a = 10$ G are measured and both field-cooled (denoted by filled markers) and zero-field-cooled (denoted by opened markers) data are shown collectively in Fig. 1. $\chi_c$ (denoted by round markers) is the molar magnetic susceptibility measured along $c$-axis and, $\chi_{ab}$ (denoted by square markers), along the tetragonal $ab$-plane. Anisotropic relation $\chi_c > \chi_{ab}$ are obvious for all field-aligned compounds below superconducting transition temperature $T_c$. Previously reported Sm$_{0.95}$La$_{0.05}$FeAsO$_{0.85}$F$_{0.15}$ ($T_c = 52$ K)[5] is also shown as a reference to $x = 0.15$. The field-aligned SmLaFeAsO$_{0.9}$F$_{0.1}$ compound is not superconducting with an antiferromagnetic Néel temperature $T_N \sim 40$ K.

Superconducting transition temperature $T_c$ were determined from the magnetic measurement data since conducting regions of aligned-powder-in-epoxy specimens are dispersed and disconnected. For near-optimum-doped $x = 0.2$ compound, both FC and ZFC data show a
sharp transition at \( T_c = 49 \) K. For optimum-doped \( x = 0.25 \) compound, a similar transition is at \( T_c = 50 \) K. For over-doped \( x = 0.3 \) specimen, a transition at \( T_c = 30 \) K was observed.

Large ZFC intragrain Meissner shielding signals were observed with \( \chi_c \sim -3 \) cm\(^3\)/mol and \( \chi_{ab} \sim -1 \) cm\(^3\)/mol for F content \( x = 0.2 \) and \( \chi_c \sim -2 \) cm\(^3\)/mol and \( \chi_{ab} \sim -0.5 \) cm\(^3\)/mol for F content \( x = 0.25 \) and 0.3 at \( T < \frac{2}{3} T_c \).

Further analysis of anisotropic diamagnetic parameter \( \gamma = \chi_c/\chi_{ab} \) of the field-aligned SmFeAs\(_{1-x}\)F\(_x\) \((x = 0.2, 0.25, 0.3)\) system in an applied magnetic field \( B_0 \) reveals that the anisotropic diamagnetic parameter \( \gamma \) are between 2.4 to 4 (Fig. 2). At \( T \sim T_c \), the FC flux-trapped signals of similar \( \gamma \) magnitude are observed. Previously reported Sm\(_{0.95}\)La\(_{0.05}\)FeAsO\(_{0.85}\)F\(_{0.15}\) is shown as a reference to \( x = 0.15 \) which has a ZFC anisotropic diamagnetic parameter \( \gamma \) of 2.4 and FC flux-trapped signal anisotropic ratio \( \gamma \) of 2.4.

Superconducting transition temperatures \( T_c \) are observed to increase with decreasing ZFC diamagnetic parameter \( \gamma \). Starting from \( T_c = 30 \) K for \( \gamma = 4 \) and \( x = 0.3 \), \( T_c \) increases to 49 K for \( \gamma = 3 \) and \( x = 0.2 \), a small increase of \( T_c \) to 50 K for \( \gamma = 2.7 \) and \( x = 0.25 \), and finally, \( T_c = 52 \) K for \( \gamma = 2.4 \) and a reference to \( x = 0.15 \) of previously reported Sm\(_{0.95}\)La\(_{0.05}\)FeAsO\(_{0.85}\)F\(_{0.15}\).

Figure 3 plots lattice aspect ratios \( c/a \) and the cell volumes against the F content of SmFeAs\(_{1-x}\)F\(_x\) \((x = 0, 0.1, 0.2, 0.25, \) and \( 0.3)\). The cell volume \( V = a^2c \) decreases linearly while the lattice aspect ratio \( c/a \) increases with F content. Since the under-doped \( x = 0.1 \) compound is antiferromagnetic while the previously reported Sm\(_{0.95}\)La\(_{0.05}\)FeAsO\(_{0.85}\)F\(_{0.15}\) (as a reference to \( x = 0.15 \) compound) is a 52 K superconductor, we suggested an antiferromagnetic and superconducting phase boundary at \( x = 0.12 \).
Figure 2. ZFC molar magnetic susceptibility ratio $\chi_c/\chi_{ab}$ in an applied magnetic field $B_a = 10$ G of SmFeAsO$_{1-x}F_x$ ($x = 0.2$, 0.25, 0.3) aligned powders. Previously reported Sm$_{0.95}$La$_{0.05}$FeAsO$_{0.85}$F$_{0.15}$ is shown as a reference to $x = 0.15$. At $T < \frac{3}{4}T_c$, $\chi_c/\chi_{ab}$ are between 2 to 4 among this system. At $T \sim T_c$, dynamic flux penetrations are obvious.

4. Conclusion

Low temperature magnetic measurements along the c-axis and ab-plane shows anisotropic relation $\chi_c > \chi_{ab}$ for both FC and ZFC modes were observed for all field-aligned specimens below superconducting transition temperature $T_c$. The under-doped compound ($x = 0.1$) is not superconducting with an antiferromagnetic Néel temperature $T_N \sim 40$ K, while the two optimum-doped compounds ($x = 0.2$ and 0.25) show high superconducting transition temperatures $T_c$ of 49K and 50K, respectively. The over-doped compound ($x = 0.3$) reveals a low superconducting temperature $T_c$ of 30 K.

The isotropic diamagnetic parameter $\gamma$ are constant and between 2.4 to 4 among this system at $T < \frac{3}{4}T_c$. In this system, superconducting transition temperatures $T_c$ were observed to raise with decreasing $\gamma$ values with the smallest value $\gamma = 2.4$ of the previously reported 52 K superconductor Sm$_{0.95}$La$_{0.05}$FeAsO$_{0.85}$F$_{0.15}$.

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