Effect of pressure on the superconducting critical temperature of La$[O_{0.89}F_{0.11}]$FeAs and Ce$[O_{0.88}F_{0.12}]$FeAs

D. A. Zocco$^a$ J. J. Hamlin$^a$ R. E. Baumbach$^a$ M. B. Maple$^a$,*
M. A. McGuire$^b$ A. S. Sefat$^b$ B. C. Sales$^b$ R. Jin$^b$
D. Mandrus$^b$ J. R. Jeffries$^c$ S. T. Weir$^c$ Y. K. Vohra$^d$

$^a$Department of Physics and Institute for Pure and Applied Physical Sciences, University of California at San Diego, La Jolla, CA 92093, USA
$^b$Materials Science & Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA
$^c$Lawrence Livermore National Laboratory, Livermore, California 94551, USA
$^d$Department of Physics, University of Alabama at Birmingham, Birmingham, Alabama 35294, USA

Abstract

We have performed several high-pressure resistivity experiments on the recently discovered superconductors La$[O_{0.89}F_{0.11}]$FeAs and Ce$[O_{0.88}F_{0.12}]$FeAs. At ambient pressure, these materials have superconducting onset temperatures $T_c$ of 28 K and 44 K, respectively. While the $T_c$ of La$[O_{0.89}F_{0.11}]$FeAs goes through a maximum between 10-68 kbar, in qualitative agreement with a recent report by Takahashi et al., the $T_c$ of Ce$[O_{0.88}F_{0.12}]$FeAs decreases monotonically over the measured pressure range. At 265 kbar, the $T_c$ of the cerium-based compound has been suppressed below 1.1 K.

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* Corresponding author.
Email address: mbmaple@ucsd.edu
1 Introduction

A new class of superconductors consisting of layered materials with the chemical formula LnOTPn, where Ln is a lanthanide element, T is a transition metal, and Pn is either P, As, or Bi, has recently emerged. The phosphorus-based versions of these compounds, LaOFeP and LaONiP have rather low superconducting critical temperatures, $T_c$ of 3 (1) and 5 K (2), respectively. Much higher $T_c$ values were achieved by fluorine-doping the corresponding arsenic-based compound to produce La[O$_{1-x}$F$_x$]FeAs, where doping to $x \sim 0.11$ produces $T_c \sim 26$ K (3). The $T_c$ appears to pass through a maximum as a function of fluorine doping. Subsequently, it was found that under a modest pressure of 40 kbar, the $T_c$ of La[O$_{0.889}$F$_{0.111}$]FeAs increases to 43 K (4), becoming the first non-cuprate superconductor with a $T_c$ higher than that of MgB$_2$. Replacing lanthanum with heavier rare-earth elements also leads to high $T_c$ values, as in Ce[O$_{1-x}$F$_x$]FeAs with $T_c$ up to 41 K (5). As of this writing, the highest $T_c$ reported for this class of materials is about 55 K, which was achieved in the compound Sm[O$_{1-x}$F$_x$]FeAs (6). The $T_c$ of optimally doped Sm[O$_{1-x}$F$_x$]FeAs initially decreases with pressure (7).

In this letter, we report measurements of $T_c$ as a function of pressure in the lanthanum- and cerium-based oxypnictide compounds to further probe the superconductivity in these interesting materials. In Ce[O$_{0.889}$F$_{0.12}$]FeAs, we found that $T_c$ decreases monotonically from 47 K (resistivity onset) to 4.5 K at 190 kbar. In La[O$_{0.889}$F$_{0.11}$]FeAs, $T_c$ appears to pass through a maximum between 10 and 68 kbar, in qualitative agreement with the results of Takahashi et al. (4).

2 La[O$_{0.89}$F$_{0.11}$]FeAs

We performed two separate high pressure-experiments on polycrystalline samples of La[O$_{0.89}$F$_{0.11}$]FeAs. The samples were taken from the same batch described in Ref. (8). The first experiment was performed in a Teflon capsule piston-cylinder cell and utilized a nearly hydrostatic 1:1 mixture of n-pentane:isoamyl alcohol as the pressure medium. Pressure was increased at room temperature and determined at low temperature using the superconducting transition of a piece of Pb located next to the sample (9). Electrical resistivity data were obtained using a 4-lead technique with a LR-700 AC resistance bridge at pressures of 0.5, 4.1 and 9.4 kbar. Figure 1 (top) shows the temperature dependence of the resistance for each pressure measured in the hydrostatic cell. Upon attempting to increase the pressure beyond 9.4 kbar, the cell failed and we continued the experiment in a diamond anvil cell (DAC).
Fig. 1. (color) Electrical resistance of La$_{0.89}$O$_{0.11}$F$_{0.11}$FeAs for different pressures. Upper panel: curves corresponding to the hydrostatic clamp. Lower panel: diamond anvil cell experiment.

Fig. 2. (color) Superconducting $T_c$ vs. pressure for La$_{0.89}$O$_{0.11}$F$_{0.11}$FeAs. Circles correspond to measurements in the hydrostatic cell, while diamonds correspond to the diamond anvil cell experiment. The dashed curve represents the data previously published by Takahashi et al. [1].

The DAC is a mechanically loaded commercial model, manufactured by Kyowa Seisakusho Ltd. The diamond anvils are beveled from 500 to 250 $\mu$m tips. One of the diamonds contains six deposited tungsten microprobes encap-
sulated in high-quality homepitaxial diamond. The fabrication of “designer” diamonds is described in Ref. (10). The gasket was made from a 200 µm thick MP35N foil preindented to 40-50 µm and a 100 µm diameter hole was drilled through the gasket using an electrical discharge machine (EDM). Several ∼ 5 µm diameter ruby spheres were loaded into the hole in the gasket and the remaining space in the hole was filled with several small chunks of polycrystalline La[O_{0.89}F_{0.11}]FeAs. Pressure was adjusted and determined at room temperature, using the fluorescence spectrum of the ruby spheres and the calibration of Chijioke et al. (11). Further details of the DAC technique are described in Ref. (12).

Figure 1 (bottom) shows the temperature dependence of the resistivity as measured in the DAC. Following the initial measurement at 68 kbar, pressure was increased to 177 kbar and then decreased to 145, 111 and 82 kbar. Note that the sample is in direct contact with the metallic gasket so that the measured resistance results from a combination of the sample and gasket resistivity. However, such a configuration is sufficient for locating the sharp drop in resistance when the sample becomes superconducting. The critical temperature was determined by the temperature at which the sample resistance reached 90% of the normal state value just above $T_c$.

Figure 2 illustrates the pressure dependence of $T_c$ for La[O_{0.89}F_{0.11}]FeAs. For comparison, the results of Takahashi et al. (4) are plotted as a dashed line. While $T_c$ initially increases and then decreases beyond ∼ 68 kbar, bearing some resemblance to the previous measurements, the magnitude of the superconducting transition temperatures are quite different. The differing pressure dependencies could be due to differences in stoichiometry or sample purity. The reported fluorine concentrations are nominal concentrations determined by the stoichiometry of the unreacted mixtures. The final concentration of fluorine in the reacted material may depend strongly on the details of the high-temperature processing such as the annealing schedule.

3 Ce[O_{0.88}F_{0.12}]FeAs

We performed two high pressure experiments on polycrystalline samples of Ce[O_{0.88}F_{0.12}]FeAs prepared at Oak Ridge National Laboratory. The compound was synthesized from CeAs, Fe$_2$O$_3$, Fe, As, and a 1:1 mixture of Ce and CeF$_3$, combined to produce a nominal composition of Ce[O_{0.88}F_{0.12}]FeAs. The reactants (3 g total mass) were intimately mixed and pressed into a pellet, sealed in an silica tube under about 1/3 atm of Ar, and heated for 36 hours at 1200 C. Reaction with vapor in the tube produces a thin light gray coating on the surface of the pellet which was gently sanded away. The sample was then re-pelletized and heated for an additional 12 hours under the same
conditions. The thin surface-layer was sanded away prior to characterization. Powder X-ray diffraction analysis revealed two small impurity phases, FeAs and CeAs, present at the level of a few percent.

At ambient pressure, the sample displays $T_c$ values of 40 and 44 K in magnetic susceptibility and resistivity, respectively. The resistive $T_c$ value is determined by the temperature at which the resistivity drops to 90% of the normal state value just above $T_c$. The first high-pressure experiment was performed in a Bridgman anvil cell with 4 mm diameter anvil flats and utilized quasi-hydrostatic solid steatite as pressure medium. Pressure is applied at room temperature using a hydraulic press and clamped with a nut. At pressures up to 44 kbar, the pressure was determined using a Sn manometer located next to the sample and the calibration of Smith et al.\textsuperscript{[13].} Above 44 kbar, the wires connecting to the Sn manometer broke and pressure was determined from the applied load. Our previous experience with these Bridgman anvil cells indicates that the pressure determined from the applied load is accurate to within ± 20%. Figure 3 shows the results of the Bridgman cell measurements on Ce[O$_{0.88}$F$_{0.12}$]FeAs.

Fig. 3. (color) Temperature dependence of the electrical resistance of Ce[O$_{0.88}$F$_{0.12}$]FeAs for different applied pressures up to 86 kbar using the Bridgman anvil cell technique. Here we have also defined the superconducting $T_c$ as the temperature at which the sample resistance reaches 90% of the normal state value. \textit{Inset:} diamond anvil cell resistance curves for 18 and 190 kbar, normalized to their respective base temperature values.

In order to verify the pressure dependence of $T_c$ and to extend the results to higher pressure, we performed a second experiment on Ce[O$_{0.88}$F$_{0.12}$]FeAs using a DAC. The technique used was the same as that described for the lanthanum-based compound. Figure 3\textit{(inset)} shows resistance versus temperature obtained from the DAC measurements. Figure 4 shows the $T_c$ versus pressure for Ce[O$_{0.88}$F$_{0.12}$]FeAs obtained from the Bridgman and DAC experi-

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ments. At the lowest measured pressures we find $T_c$ values near 47 K, somewhat higher than the $T_c = 44$ K measured at ambient pressure. This small increase in the critical temperature is likely due to improvement in sample connectivity due to grain compaction under pressure, rather than the intrinsic pressure dependence of $T_c$. The $T_c$ value decreases monotonically with pressure and is suppressed below 1.1 K at 265 kbar.

The strong dependence of $T_c$ on pressure in these materials is rather remarkable. Shein et al. \cite{14} calculate that the bulk modulus of LaOFeAs is only 98 GPa, significantly smaller than that found for the cuprate superconductors. It is likely that the strong dependence of $T_c$ on pressure for La$_{1-x}$FeAs and Ce$_{1-x}$F$_{1.12}$FeAs is related to their high compressibility. Experiments to determine structural parameters under pressure would help to clarify the effect of lattice properties on $T_c$.

For the oxypnictides, it is likely that, as in the cuprates, increasing pressure leads to an increase in carrier concentration. The initial increase in $T_c$ with pressure for La$_{1-x}$FeAs may thus be due to the sample being underdoped. Indeed, Lu et al. \cite{15} find that increased doping achieved through high-pressure synthesis raises $T_c$ to 41 K in La$_{0.4}$FeAs. In the high-$T_c$ cuprate superconductors, it is found that $T_c$ generally increases with pressure in optimally doped samples, highlighting the fact that the effect of pressure is more complicated than simply changing the carrier concentration \cite{16}. The negative pressure dependence of $T_c$ that we find for apparently optimally doped Ce$_{1-x}$F$_x$FeAs combined with that previously reported for Sm$_{1-x}$F$_x$FeAs \cite{7} points to a possible difference between the oxypnictide and cuprate superconductors. A systematic study of the effect of pressure on $T_c$ across a wide range of dopings is clearly needed in order to obtain a better understanding of the optimal conditions for high-$T_c$ values in the oxypnictide superconductors.

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Fig. 4. (color) Superconducting $T_c$ versus pressure for Ce[O$_{0.88}$F$_{0.12}$]FeAs. The dashed curve is a guide to the eye given by a quadratic fit of the data, extrapolating to zero temperature at $\sim$ 190 kbar.

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