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Specific heat of the iron-based high-T_c superconductor $SmO_{1-x}F_xFeAs$

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The specific heat $C(T)$ of iron-based high-T_c superconductor $SmO_{1-x}F_xFeAs$ $(0 \leq x \leq 0.2)$ was systematically studied. For the undoped $x=0$ sample, a specific heat jump was observed at 130 K. This is attributed to the structural or spin-density-wave transition, which also manifests on resistivity as a rapid drop. However, this jump disappears with slight F doping in the $x=0.05$ sample, although the resistivity drop still exists. The specific heat C/T shows clear anomaly near T_c for $x=0.15$ and 0.20 superconducting samples. Such anomaly has been absent in $LaO_{1-x}F_xFeAs$. For the parent compound SmOFeAs, $C(T)$ shows a sharp peak at 4.6 K, and with electron doping in the $x=0.15$ sample, this peak shifts to 3.7 K. It is interpreted that such a sharp peak results from the antiferromagnetic ordering of Sm^{3+} ions in this system, which mimics the electron-doped high-T_c cuprate $Sm_{2-x}Ce_xCuO_{4-\delta}$.

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The recent discovery of superconductivity at $T_c=26$ K in iron-based $LaO_{1-x}F_xFeAs$ $(x=0.05-0.12)$ (Ref. 1) has attracted great attention. Following this initial work, more compounds with T_c as high as 55 K were synthesized by replacing La with other rare-earth elements such as Sm,[2–4] Ce,[5] Nd,[6] Pr,[7] and Gd.[8] Due to their high superconducting transition temperature, which is second only to the high-T_c cuprate superconductors, enormous experimental and theoretical efforts have been put on these materials to clarify their phase diagram and superconducting mechanism.

These quaternary rare-earth transition metal arsenide oxides LnOFeAs (Ln=La, Sm, Ce, Nd, Pr, and Gd) form a tetragonal ZrCuSiAs-type structure.[9] It is believed that the Fe-As layers are responsible for the superconductivity and Ln-O layers provide electron carriers through fluorine doping, or very recently by simply introducing oxygen vacancies.[10] Neutron scattering experiments have demonstrated that the undoped parent compound LaOFeAs develops long-range spin-density-wave (SDW)-type antiferromagnetic order below 150 K.[11,12] With increasing electron doping by fluorine, the SDW order is suppressed and superconductivity emerges, suggesting competing orders in these systems and similar phase diagram to the one in high-T_c cuprates.[4,13] Theoretically, electron-phonon coupling is not sufficient to explain superconductivity in $LaO_{1-x}F_xFeAs$,[14] while antiferromagnetic interaction[15–17] and Hund's rule ferromagnetic interaction[18,19] have been considered as the possible pairing mechanism.

Among the family of $LnO_{1-x}F_xFeAs$, specific heat was only studied for $LaO_{1-x}F_xFeAs$ compounds so far.[12,13,20,21] Clear specific heat jump was observed at the temperature about 150 K for LaOFeAs,[12,13] which is accompanied by anomalies on resistivity, Hall coefficient, and Seebeck coefficient.[12,13] Since structural transition was also found at 150 K by neutron scattering[11] and x-ray diffraction,[22] at present it is unclear whether these resistivity and specific heat anomalies around 150 K are due to the structural or SDW transition, or to both. For superconducting $LaO_{0.9}F_{0.1-\delta}FeAs$ with $T_c \approx 28$ K, a nonlinear magnetic field dependence of the electronic specific heat coefficient $\gamma(H)$ has been found in the low-temperature limit, which is consistent with the prediction for a nodal superconductor and suggests an unconventional mechanism for this superconductor.[20] However, it is surprising that no visible specific heat anomaly was detected near T_c on the raw data for superconducting $LaO_{1-x}F_xFeAs$ samples despite the large Meissner fractions,[20,21] and only a broadened anomaly of $[C(0T)\text{-}C(9T)]/T$ was observed.[20] This result may reflect the low superfluid density in $LaO_{1-x}F_xFeAs$. For comparison, precise specific heat measurements on other compounds of this family are highly desired.

Here we systematically study the specific heat of $SmO_{1-x}F_xFeAs$ for $0 \leq x \leq 0.2$, with the maximum T_c(onset)=54 K at $x=0.2$. A specific heat jump was observed at 130 K for the undoped $x=0$ sample, indicating the structural or SDW transition. However, this jump disappears in the $x=0.05$ sample. The specific heat C/T shows clear anomaly near T_c for $x=0.15$ and 0.20 superconducting samples, which has been absent in $LaO_{1-x}F_xFeAs$. The sharp peak of $C(T)$ appears at 4.6 K for the parent compound SmOFeAs, and it shifts to 3.7 K with doping electrons in the $x=0.15$ sample. This specific heat peak should come from the antiferromagnetic ordering of Sm^{3+} ions in this system, as in the electron-doped high-T_c cuprate $Sm_{2-x}Ce_xCuO_{4-\delta}$.

The polycrystalline samples with nominal composition $SmO_{1-x}F_xFeAs$ ($x=0$, 0.05, 0.15, and 0.20) are the same ones as studied in Ref. 4, synthesized by conventional solid-state reaction. The $x=0$ and 0.05 samples are in single phase. A trace of impurity phases SmOF and SmAs can be observed in $x=0.15$ sample, and these impurities are estimated to be less than 10% in the $x=0.20$ sample. Specific heat measurements were performed in a Quantum Design physical property measurement system (PPMS) via the relaxation method and the results are presented per mole of atom (J/mol K). Magnetic field $H=8$ T was applied for the $x=0.15$ and 0.20 superconducting samples.

The resistivity of this series of $SmO_{1-x}F_xFeAs$ samples have been reported previously.[4] For the $x=0$ sample, a rapid resistivity drop below about 130 K was observed. Superconductivity emerges at $x=0.10$, and the $x=0.20$ sample has the

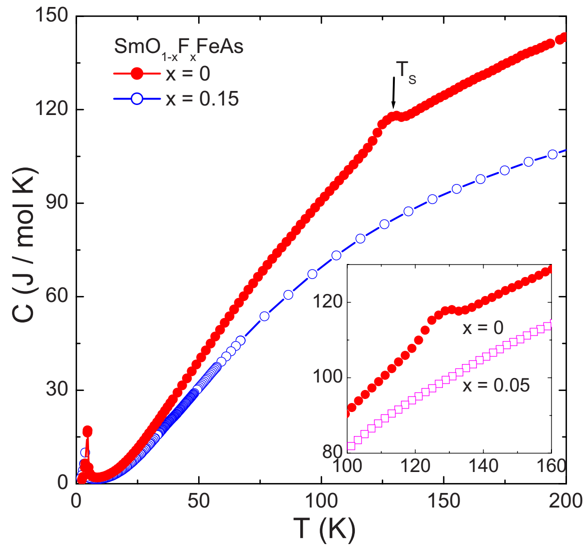

FIG. 1. (Color online) Specific heat of $SmO_{1-x}F_xFeAs$ samples with $x=0$ and 0.15. For the $x=0$ parent compound, clear specific heat jump can be seen at 130 K, denoted as the structural or SDW transition temperature T_s. The inset shows the lack of such jump in the slightly F-doped $x=0.05$ sample.

maximum superconducting transition temperature T_c(onset) $=54$ K. The superconducting volume fractions of the $x=0.20$ sample at 5 K were estimated to be 60% and 30% from the susceptibility measured under zero-field-cool and field-cool conditions at 10 Oe.[4]

Figure 1 shows the specific heat $C(T)$ of $SmO_{1-x}F_xFeAs$ samples with $x=0$ and 0.15. There is a clear specific heat jump close to 130 K for the $x=0$ sample (enlarged in the inset). A similar jump has been observed in LaOFeAs at 150 K.[12,13] The specific heat jump at 130 K of the SmOFeAs sample is consistent with the resistivity drop.[4] As in LaOFeAs, the specific heat and resistivity anomalies in SmOFeAs are also attributed to a structural or SDW transition. We note that the phase transition temperature T_s in SmOFeAs is about 20 K lower than that in LaOFeAs. The inset of Fig. 1 shows the lack of specific heat jump in the slightly F-doped $x=0.05$ sample, although there is still a resistivity drop (but less sharp) at about 110 K.[4] This result suggests that electron doping suppresses the magnetic order and structural distortion in SmOFeAs. Indeed, the static antiferromagnetic SDW order and structural transition disappear in the doped superconducting $LaO_{1-x}F_xFeAs$ samples, shown by neutron scattering and x-ray diffraction experiments.[11,22] At low temperature there is a sharp peak for both $x=0$ (nonsuperconducting) and 0.15 (superconducting) samples, which will be discussed later.

Figure 2(a) plots C/T vs T for the optimally doped $SmO_{0.80}F_{0.20}FeAs$ sample in zero and $H=8$ T magnetic fields. Below the zero resistivity $T_c=50$ K, one can see a clear specific heat anomaly. Such anomaly has been absent in the $LaO_{1-x}F_xFeAs$ superconducting samples with large Meissner fractions.[20,21] The difference may reflect the higher superfluid density in $SmO_{1-x}F_xFeAs$. This is reasonable, since the maximum T_c of $SmO_{1-x}F_xFeAs$ is twice that of $LaO_{1-x}F_xFeAs$.

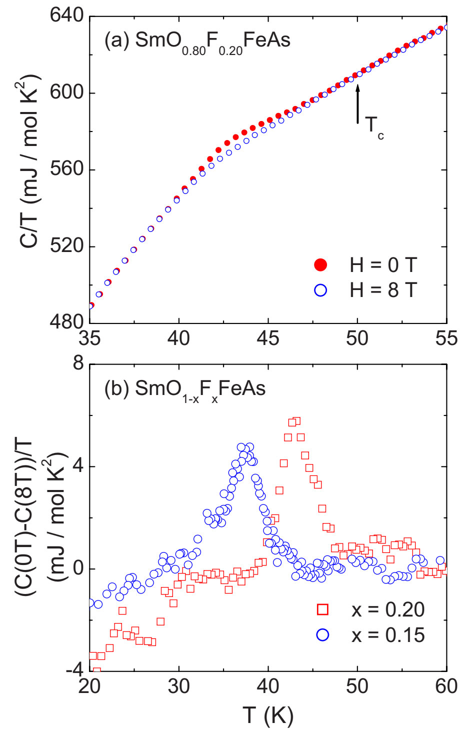

FIG. 2. (Color online) (a) Specific heat of optimally doped $SmO_{0.80}F_{0.20}FeAs$ sample in zero and $H=8$ T magnetic fields, plotted as C/T vs T close to T_c. The arrow marks zero resistivity $T_c=50$ K. (b) $[C(0T)-C(8T)]/T$ vs T for the $x=0.15$ and 0.20 samples. One can see a clear specific heat peak near the zero resistivity T_c for both samples.

Although 8 T is far away from the upper critical field H_{c2} which is higher than 60 T,[23] we nevertheless plot $[C(0T)-C(8T)]/T$ vs T for the $x=0.15$ and 0.20 samples in Fig. 2(b). The obtained specific heat peak near the zero resistivity T_c is sharper than that in the $LaO_{0.9}F_{0.1-\delta}FeAs$ sample.[20]

Below we focus on the low-temperature specific heat behavior of $SmO_{1-x}F_xFeAs$ at $T<20$ K. In Fig. 3(a), $C(T)$ of the $x=0$ sample shows a very sharp peak at 4.6 K. With electron doping in the $x=0.15$ superconducting sample, the peak shifts to 3.7 K and its height decreases. This low-temperature peak has not been seen in previous specific heat studies of $LaO_{1-x}F_xFeAs$.[12,13,20,21] Since the only difference between these two materials is the Ln^{3+} ions, i.e., nonmagnetic La^{3+} and magnetic Sm^{3+} ions, this peak may relate to the magnetic ordering of Sm^{3+} ions. In fact, exactly the same specific heat behavior at low temperature has been observed in electron-doped high-T_c cuprate $Sm_{2-x}Ce_xCuO_{4-\delta}$.[24,25] Antiferromagnetic ordering of the Sm^{3+} ions was found in Sm_2CuO_4 at $T_N=5.9$ K, accompanied by a sharp specific heat peak.[24] By substituting electron donor element Ce^{4+} for Sm^{3+} ions, the ordering temperature T_N is lowered to 4.7 K in $Sm_{1.85}Ce_{0.15}CuO_{4-\delta}$ with $T_c=16.5$ K.[25] Therefore the sharp specific heat peak below 5 K in $SmO_{1-x}F_xFeAs$ manifests the antiferromagnetic ordering of Sm^{3+} ions. Below T_N, the superconductivity coexists with antiferromagnetism, as in $Sm_{2-x}Ce_xCuO_{4-\delta}$.

Due to this antiferromagnetic specific heat peak, it is not

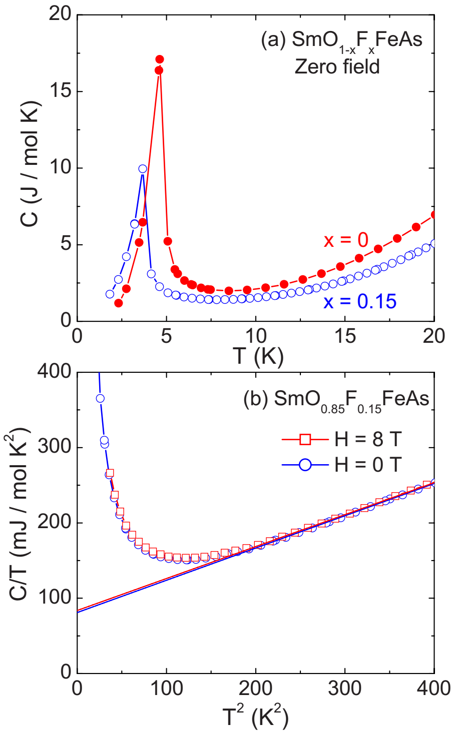

FIG. 3. (Color online) (a) Low-temperature specific heat of $SmO_{1-x}F_xFeAs$ samples with $x=0$ and 0.15 in zero field. The sharp peak comes from the antiferromagnetic ordering of Sm^{3+} ions in this system. (b) C/T vs T^2 for the $x=0.15$ sample in $H=0$ and 8 T. The lines are linear fits between 14 and 20 K.

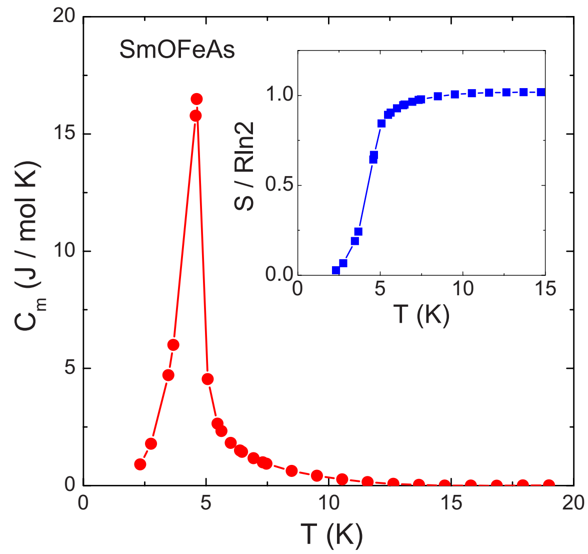

FIG. 4. (Color online) Magnetic specific heat of SmOFeAs, $C_m=C-\gamma T-\beta T^3$. Inset: Entropy associated with the magnetic transition.

easy to extrapolate the electronic specific heat coefficient γ in the low-temperature limit. In Fig. 3(b), C/T vs T^2 is plotted for the $x=0.15$ sample in $H=0$ and 8 T. The data between 14 and 20 K can be linearly fitted by $C/T=\gamma+\beta T^2$, which give $\gamma=81.0$ and 83.7 mJ/mol K^2 for $H=0$ and 8 T, respectively. This value of γ is much higher than that obtained in $LaO_{0.89}F_{0.11}FeAs$ ($T_c \approx 28$ K), $\gamma=1.0$ mJ/mol K^2 (Ref. 21). For $Sm_{1.85}Ce_{0.15}CuO_{4-\delta}$ with $T_c=16.5$ K, the fitting also gave exceptionally large $\gamma=103.2$ mJ/mol K^2 (Ref. 25). It was speculated that the effects of magnetic correlation exist well above T_N, thereby making accurate determination of γ difficult. In addition, since the fitting in Fig. 3(b) was done at relatively high temperature and over a small range from 14 to 20 K, the slope β may not represent the phonon specific heat in the low-temperature limit, where it is proportional to T^3. Therefore the resulting large γ may not be reliable. In Fig. 3(b), γ only increases very slightly in $H=8$ T and we are unable to examine its field dependence for the $SmO_{0.85}F_{0.15}FeAs$ sample. For superconducting $LaO_{1-x}F_xFeAs$, there is no such antiferromagnetic specific heat peak and the data were fitted at low temperature, thus the extrapolated $\gamma \sim 1.0$ mJ/mol K^2 is more reliable and shows a steep increase with increasing magnetic field.[20,21]

Figure 4 plots the magnetic specific heat of SmOFeAs, $C_m=C-\gamma T-\beta T^3$, with $\gamma=119.4$ mJ/mol K^2 and $\beta=0.56$ mJ/mol K^4 obtained from the same fitting process as in Fig. 3(b). The magnetic entropy S associated with the magnetic transition is also calculated from the $C_m(T)$ and shown in the inset of Fig. 4. With increasing temperature, the entropy rapidly increases and then saturates to the value of $R \ln 2$ within experimental error, indicating that the Sm^{3+} ground state in the crystal field is a doublet for SmOFeAs.

In summary, we have systematically studied the specific heat of the iron-based high-T_c superconductor $SmO_{1-x}F_xFeAs$. First, a specific heat jump was observed at 130 K for the undoped $x=0$ sample, corresponding to the structural or SDW transition. However, this jump disappears in the slightly F-doped $x=0.05$ sample, indicating the suppression of the SDW order and structural distortion by electron doping in this system. Second, a clear specific heat anomaly can be seen near T_c for superconducting $SmO_{1-x}F_xFeAs$ samples, while it has been absent in $LaO_{1-x}F_xFeAs$. This result suggests higher superfluid density in $SmO_{1-x}F_xFeAs$. Finally, sharp specific heat peak shows up at 4.6 K for the $x=0$ sample, and it shifts to 3.7 K upon electron doping in the $x=0.15$ sample. By comparing with the electron-doped high-T_c cuprate $Sm_{2-x}Ce_xCuO_{4-\delta}$, this sharp peak is attributed to the antiferromagnetic ordering of Sm^{3+} ions in the $SmO_{1-x}F_xFeAs$ system.

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