Superconducting Gap and Pseudogap in Sm(O$_{1-x}$F$_x$)FeAs Layered Superconductor from Photoemission Spectroscopy

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(Dated: May 25, 2008)

High resolution photoemission measurements have been carried out on non-superconducting SmOFeAs parent compound and superconducting Sm(O$_{1-x}$F$_x$)FeAs ($x=0.12$, and 0.15) compounds. The momentum-integrated spectra exhibit a clear Fermi cutoff that shows little leading-edge shift in the superconducting state which suggests the Fermi surface sheet(s) around the $\Gamma$ point may not be gapped in this multiband superconductors. A robust feature at 13 meV is identified in all these samples. Spectral weight suppression near $E_F$ with decreasing temperature is observed in both undoped and doped samples that points to a possible existence of a pseudogap in these Fe-based compounds.

PACS numbers: 74.70.-b, 74.25.Jb, 79.60.-i, 71.20.-b

The recent discovery of superconductivity in iron-based oxyynictides has attracted much attention because of their unusual high superconducting transition temperature ($T_c$). The highest $T_c$ achieved so far (55 K) has exceeded the generally-believed limit ($\sim 40$ K) set by the traditional BCS theory of superconductivity, putting them into the second class of "high temperature superconductors" in addition to the cuprates. The parent compound of these iron-based superconductors has been found to exhibit a spin density wave(SDW)-like transition near 150 K and a possible antiferromagnetic ground state. Doping charge carriers into the system induces superconductivity at appropriate doping levels. These behaviors appear to be similar to those in cuprate superconductors. One natural question to ask is whether the superconductivity mechanism in these new Fe-based superconductors is unconventional, and whether it may share the same mechanism as that in the cuprate superconductors.

There are a couple of related issues that need to be addressed about these new Fe-based superconductors. One is about the ground state of undoped parent compound and how strong the electron correlation is. The second is about the superconducting gap symmetry and the pairing mechanism. The third is on whether its normal state is anomalous and whether there is a pseudogap in the normal state as found in the cuprate superconductors. Photoemission spectroscopy, as a powerful tool to directly measure the electronic structure and energy gap, can shed important light on these issues.

In this paper, we report high resolution photoemission measurements on the Sm(O$_{1-x}$F$_x$)FeAs compounds at various doping levels. We have found that the momentum-integrated spectra exhibit a clear Fermi cutoff with little leading-edge shift in the superconducting state. This suggests that, in this multiband superconductor, different Fermi surface sheets may have different superconducting gaps and the Fermi surface sheet(s) around the $\Gamma$ point may not be gapped. We have also identified a robust feature at 13 meV that is present in all these samples with different doping levels. Near-$E_F$ spectral weight suppression with decreasing temperature is observed in both undoped and doped samples. It points to a possible existence of a pseudogap in these Fe-based compounds.

The photoemission measurements have been carried out on our newly-developed system using both Vacuum Ultraviolet (VUV) laser and Helium discharge lamp as light sources. The advantages of the VUV laser photoemission lie in its super-high energy resolution, bulk sensitivity and high photon flux to give high statistics of the data. The photon energy of the VUV laser is 6.994 eV with a bandwidth of 0.26 meV. For the laser measurements, the energy resolution of the electron energy analyzer (Sciencta R4000) was set at 1.5 meV, giving an overall energy resolution of 1.52 meV. The spot size of the laser is less than 0.2 mm. The Helium lamp can provide two photon energies at 21.218 eV (Helium I resonance line) and 40.813 eV (Helium II resonance line). The energy resolution for the 21.218 eV measurements
was set at 7.5 meV while for the 40.813 eV measurements at 20~30 meV.

The polycrystalline Sm(O₁₋ₓFx)FeAs (x=0, 0.12 and 0.15, nominal composition) samples are prepared by solid state reaction method⁴,⁸,⁹. The x=0 sample is not superconducting but with a possible SDW transition at ~150 K, while the x=0.12 and 0.15 samples are superconducting with Tc at 26 K and 43 K, respectively⁹. For a polycrystalline sample, in order to probe its intrinsic electronic structure through photoemission measurements, great care has to be taken to obtain clean surface and minimize the sample aging effect. To get clean surface, we tried both scraping with a diamond file and fracturing. We found that fracturing leads to cleaner surface, as judged by the intensity and sharpness of the characteristic -0.2 eV feature in the valence band (Fig. 1). Therefore, all samples in this work were measured by fracturing in situ in vacuum with a base pressure better than 5×10⁻¹¹ Torr. In addition, we found that these samples show aging effect even in the ultra-high vacuum, manifested by depletion of the near-E_F spectral weight and particularly the change with time of the high binding energy part in VUV laser photoemission spectra. Therefore, all the laser photoemission data presented in the work were measured on a fresh sample surface within 3~4 hours after sample fracturing.

Fig. 1 shows valence band of Sm(O₁₋ₓFx)FeAs with different doping levels (x=0, 0.12) and at different temperatures measured using different photon energies of the Helium lamp. The main feature in these compounds is the peak near -0.2 eV¹⁷. Different photon energy gives similar spectra, as seen from the 21.2 eV and 40.8 eV measurements on the x=0 sample. There is little spectral change with temperature except for the near-E_F region, as shown in the inset of Fig. 1. There is no obvious valence band change observed between different samples (x=0 and 0.12), specifically, the -0.2 eV peak shows little position shift with doping.

Fig. 2 shows photoemission spectra of Sm(O₁₋ₓFx)FeAs (x=0, 0.12, and 0.15) measured using VUV laser at 13 K, together with data taken at 15 K using 21.218 eV photon energy from the Helium lamp. Two obvious kink features can be identified from these...
data. One is at $\sim13$ meV where the spectrum starts to deviate from the linear behavior at high binding energy. The other is at lower binding energy near 4 meV that is due to the Fermi function cutoff. The super-high energy resolution and low temperature make the 13 meV feature well separated from the Fermi cutoff and clearly visible. We note that this 13 meV feature is robust because it is present in both undoped sample and doped samples, and seen in both laser photoemission data and high resolution helium lamp measured data.

The high resolution and low temperature data in Fig. 2 make it possible to examine possible superconducting gap in the $x=0.12$ and $x=0.15$ superconductors. Generally speaking, in a momentum-integrated photoemission spectrum, an $s$-wave superconducting gap on a single Fermi surface can be easily identified from the leading-edge shift, as demonstrated in Boron-doped superconducting diamond where the leading-edge shift is visible even for a superconducting gap less than 1 meV\textsuperscript{18}. But for a system with multiple superconducting gaps on different Fermi surface sheets, the situation becomes not so straightforward. The leading edge position in this case is mainly dictated by the minimum superconducting gap on a Fermi surface sheet. The larger energy gaps on the other Fermi surface sheets will then show up as additional features in the spectrum at higher binding energy. For a superconductor with two $s$-wave gaps like MgB$_2$\textsuperscript{12}, both the leading edge shift and high binding energy feature are resolved to represent the multiple gap structures. The situation will get more complicated if non-$s$-wave gaps are involved on different Fermi surface sheets and/or superconducting coherence peaks are not well developed.

The Fe-based compounds have multiple Fermi surface sheets around $\Gamma$ point and $M(\pi, \pi)$ point, as shown in the inset of Fig. 2\textsuperscript{11}. It is possible that these Fermi surface sheets may have different superconducting order parameters. As seen in Fig. 2, there is no obvious coherence peaks developed on the spectra in the superconducting state, even for the $x=0.15$ sample with $T_c$ as high as 43 K. This lack of coherence may come from disorder or unconventional pairing symmetry. Particularly, the leading edge of the spectra shows minimal shift from the Fermi level, as seen from the data in Fig. 2 for both $x=0.12$ sample with $T_c=26$ K and $x=0.15$ sample with $T_c=43$ K. In fact, the Fermi cutoff for these samples can be well fitted with a Fermi-Dirac distribution function convoluted with the energy resolution used (Fig. 2), indicating a nearly zero leading-edge shift. Supposing this zero-shift Fermi edge is not from non-superconducting metallic impurities in the samples, it indicates that there are ungapped Fermi surface sheet(s) in the Sm($O_{1-x}F_x$)FeAs superconductors. We note that with a laser photon energy at 6.994 eV, the momentum space it can cover does not span the entire Brillouin zone: it covers the Fermi surface sheets around $\Gamma$ point, but not those around the $M(\pi, \pi)$ point (inset of Fig. 2). The laser data then further suggest that there are ungapped Fermi surface sheet(s) near the $\Gamma$ point.

It is tempting to see whether the $\sim13$ meV feature might represent a superconducting gap in the
Sm(O$_{1-x}$F$_x$)FeAs superconductors. However, this possibility can be simply ruled out because it is also observed in the undoped x=0 sample that is not superconducting. We do not see other obvious features from the laser data between the Fermi level and 13 meV (Fig. 2) that can be taken as signatures of the superconducting gap on other Fermi surface sheets. To probe the superconducting gap on the Fermi surface sheets near the M($\pi$, $\pi$) point (inset of Fig. 2), one can in principle rely on high photon energy data that can cover the entire Brillouin zone, like the Helium lamp data in the inset of Fig. 1. However, given that the Fermi surface sheet(s) around the $\Gamma$ point is not gapped in the superconducting state as suggested from our laser data, it will also give rise to a zero-shifted leading edge in the high photon energy data. This would make it difficult to extract the gap information from the leading edge in the high photon energy spectrum. Again, one has to look for signatures at higher binding energy in the Helium lamp (or other high photon energy) spectra that require high energy resolution and high data statistics.

Fig. 3 shows detailed temperature dependence of the laser photoemission spectra for the undoped SmOF$_x$As sample. The temperature-induced change is mainly confined near the Fermi level region; the high binding energy spectra at different temperatures can be normalized to overlap with each other. Note that the spectra at different temperatures do not cross the same point at the Fermi level, a behavior that is different from a normal metal like gold where all spectra cross at the same energy $E_F$. To remove the effect of Fermi cutoff, the spectra are divided by Fermi-Dirac distribution function at the respective temperature and shown in the inset of Fig. 3. One can see a suppression of the spectral weight near the Fermi level with decreasing temperature, a behavior that starts at high temperatures even at 150 K. This behavior is similar to the normal state spectral weight depletion near the antinodal region in the underdoped cuprate superconductors which is related to the opening of a pseudogap[14].

To further examine the possible opening of the pseudogap in Fe-based compounds, we follow the procedure that is commonly used in high-$T_c$ cuprate superconductors[19] by symmetrizing the original data in Fig. 3a with respect to the Fermi level (Fig. 3b). This is another way to remove the Fermi-Dirac distribution function and it provides a visualized way to look for a gap. Again one sees clearly the depletion of spectral weight near the Fermi level with decreasing temperature (Fig. 3b). To highlight the effect caused by temperature, we further divide the symmetrized spectra with the one at 200 K (Fig. 3c). The suppression of the spectral weight near the Fermi level becomes clearer and one can now identify an energy scale at which the spectral weight starts to lose. It is $\sim$25 meV for the 13 K data, shows slight increase with increasing temperature, but overall lies in the 25$\sim$40 meV energy range for different temperatures (Fig. 3c).

The temperature dependence of spectra for the x=0.12 superconducting sample ($T_c=26$ K) (Fig. 4) appears to be surprisingly similar to that in the undoped SmOF$_x$As (Fig. 3). Here again one sees suppression of spectral weight with decreasing temperature as in the inset of Fig. 4a, symmetrized data in Fig. 4b and normalized data in Fig. 4c. For this superconducting sample with $T_c=26$ K, one may wonder whether the near-$E_F$ spectral weight suppression in the superconducting state, like the 13 K data in Fig. 4, compared with a normal state data at 50 K, can be taken as a signature of superconducting gap opening. Note that the same behavior also occurs in the non-superconducting SmOF$_x$As sample (Fig. 3), we believe this is not a reliable way in judging on a superconducting gap.

In summary, from our high resolution photoemission measurements on the Sm(O$_{1-x}$F$_x$)FeAs compounds, we found zero leading-edge shift in the spectra in the superconducting state. This suggests that the Fermi surface sheet(s) around the $\Gamma$ point may not be gapped. We have identified a robust feature at 13 meV in different samples. Whether this could be caused by electron coupling with some bosonic mode needs to be further investigated. Spectral suppression near $E_F$ with decreasing temperature is observed in both undoped and doped samples. This points to possible existence of a pseudogap in the Fe-based compounds. Whether this is caused by local SDW fluctuation or strong electron-boson coupling needs further experimental and theoretical studies.

This work is supported by the NSFC, the MOST of China (973 project No: 2006CB601002, 2006CB921302), and CAS (Projects ITSNEM).

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[1] Y. Kamihara et al., J. Am. Chem. Soc. 130, 3296 (2008).
[2] G. F. Chen et al., arXiv:cond-mat/0803.0128.
[3] H. H. Wen et al., Europhys. Lett. 82, 17009 (2008).
[4] X. H. Chen et al., arXiv:cond-mat/0803.3603.
[5] G. F. Chen et al., arXiv:cond-mat/0803.3790.
[6] Z. A. Ren et al., Europhys. Lett. 82, 57002 (2008).
[7] Z. A. Ren et al., arXiv:cond-mat/0803.4283.
[8] Z. A. Ren et al., Chin. Phys. Lett. 25, 2215 (2008).
[9] R. H. Liu et al., arXiv:cond-mat/0804.2105.
[10] J. G. Bednorz et al., Z. Phys. B 64, 189 (1986).
[11] J. Dong et al., arXiv:cond-mat/0803.4262.
[12] C. Cruz et al., arXiv:cond-mat/0804.0795.
[13] M. A. McGuire et al., arXiv:cond-mat/0804.0796.
[14] T. Timusk and B. Statt, Rep. Prog. Phys. 62, 61 (1999).
[15] A. Damschel et al., Rev. Mod. Phys. 75, 473 (2003).
[16] G. D Liu et al., Rev. Sci. Instruments 79, 023105 (2008).
[17] H. W. Ou et al., arXiv:cond-mat/0803.4328.
[18] K. Ishizaka et al., Phys. Rev. Lett. 98, 047003 (2007).
[19] S. Tsuda et al., Phys. Rev. B 72, 064527 (2005).
[20] M. R. Norman et al., Phys. Rev. B 57, R11093 (1998).