Transport and infrared properties of SmFeAs(O$_{1-x}$F$_x$): from SDW to superconducting ordering

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Abstract
We report measurements of resistivity, magnetoresistivity, Hall effect, Seebeck coefficient and infrared reflectivity of undoped SmFeAsO and lightly doped SmFeAs(O$_{0.93}$F$_{0.07}$) oxypnictides. All the properties measured on SmFeAsO are characterized by clear signatures of the magnetic instability. A self-consistent picture emerges in which carrier condensation occurs below the magnetic transition, due to the opening of a spin density wave (SDW) gap. This is accompanied by the mobility increase of not-gapped carriers due to the suppression of electron–electron scattering. SmFeAs(O$_{0.93}$F$_{0.07}$) exhibits an increase of the metallic character on cooling consistent with electron doping, even though at room temperature values of all the properties nearly overlap with those of SmFeAsO. However, with a decrease in temperature all anomalies related to the SDW instability are missing and the superconducting transition occurs. This suggests that doping abruptly breaks the symmetries of the Fermi surface, inhibiting the SDW formation in favor of the superconducting transition, with no substantial changes in the density of states or in the effective mass.

(Some figures in this article are in colour only in the electronic version)

1. Introduction
The recent discovery of superconductivity at 26 K in LaFeAsO$_{1-x}$F$_x$ [1] attracted a lot of attention to rare earth (RE) oxypnictides. These phases crystallize in the tetragonal system at room temperature and their structure is built up by two kinds of planar layers constituted of edge sharing tetrahedra stacked along the c axis. The former layer is constituted of tetrahedra centered by O with the RE at vertices (charge reservoir layer), whereas in the latter Fe coordinates As (conducting layer). The parent compound exhibits an antiferromagnetic (AF) transition around 140–150 K [1–6] attributed to the development of a spin density wave (SDW) with a small moment in the Fe–As plane, as indicated by neutron diffraction analysis [3] and by Mössbauer spectroscopy (magnetic measurements) [7]. The magnetic transition has been characterized [2, 8, 9] and related to a tetragonal–orthorhombic structural transition detected by different techniques [3, 10–13]. In LaFeAsO the neutron diffraction studies have pointed out well-separated structural and magnetic transitions [3, 7], with the former one occurring at a slightly higher temperature. In SmFeAsO detailed x-ray analysis has characterized the structural transition [11, 12], while direct evidence of AFM ordering is still lacking.

Electron doping suppresses the magnetic instability in favor of superconductivity and a critical temperature as high as 55 K in the SmFeAs(O$_{1-x}$F$_x$) has been obtained [14].
The layered structure and the rather high critical temperature, hardly explainable by the electron–phonon coupling, seems to suggest a similarity with high temperature superconductors. However, different from cuprates, many experiments suggest a multiband nature of superconductivity in these compounds [15, 16] as in the case of magnesium diboride.

For the better comprehension of the superconducting mechanism in oxypnictides, a systematic study of electrical and thermo-electrical transport properties and of their dependence upon doping in single crystals would be highly desirable. Unfortunately, up to now, only small single crystals are available [17] and such kind of analysis has been performed mainly on polycrystalline samples [18–20], especially by comparison of magnetoresistivity and the Hall effect.

In this work we report resistivity, Hall effect, magnetoresistivity, Seebeck coefficient and infrared (IR) conductivity measurements on SmFeAsO and SmFeAs(0.93F0.07). The undoped sample exhibits a clear anomaly at •130–140 K. Although the nature of this anomaly is not yet definitively explained, in the following we refer to it as due to the occurrence of an SDW ordered state. The same properties measured on SmFeAs(O0.93F0.07) show that the low level of doping does not substantially modify phonon and electron parameters, yet it completely suppresses the SDW transition in turn of superconductivity. Concerning the superconducting state, IR reflectivity data show two different spectral features. The former is related to the gap in the ab plane, the latter to an interplane Josephson coupling. These features are similar to those observed in high-\(T_c\) cuprates.

### 2. Experimental details

Samples with nominal compositions SmFeAsO and SmFeAs(\(O_{0.93}F_{0.07}\)) were prepared in three steps as reported in [21]:

1. First, heating Sm and As in an evacuated glass flask at a maximum temperature of 550 \(\degree\)C to synthesize SmAs, and then reacting the arsenide with stoichiometric amounts of Fe, \(Fe_2O_3\) and \(FeF_2\) in the form of a pellet at 1200 \(\degree\)C for 24 h in an evacuated quartz flask. Finally, the products underwent a further sintering step at 1300 \(\degree\)C for 72 h in an evacuated quartz flask in order to obtain a compact sample suitable for transport measurements. The effect of sintering is to increase the density and connection between the grains, improving substantially their transport properties.

2. Phase identification was performed by XRPD (Philips PW1830; Bragg–Brentano geometry; Cu K\(\alpha_1\), \(\alpha_2\); range 20–110° 2\(\theta\); step 0.025° 2\(\theta\); sampling time 12 s) and structural refinement was successfully carried out in the space group \(P4/nnm\) according to the Rietveld method using the program Fullprof. The samples were characterized also by scanning electron microscope (SEM) and transmission electron microscope (TEM) analyses. By means of synchrotron powder diffraction, coupled with Rietveld refinement, an orthorhombic to monoclinic phase transition has been ascertained in SmFeAsO by measurements done at room temperature and 100 K, respectively [11].

Rietveld refinement of x-ray diffraction patterns and SEM observation reveal the single-phase nature of the samples, whereas SEM-EDS analyses are in good agreement with the expected nominal SmFeAsO composition. No evidence for nanodomains, twinning, extended defects or superlattice reflections can be obtained by TEM observation, thus suggesting a high degree of crystallinity of our samples [21]. The effect of sintering is to increase the density and connection between the grains, improving substantially their transport properties. The good quality of the samples is proved also by thermal [8] and magnetization [21] measurements, which are reported elsewhere.

Magnetoresistivity, the Hall effect and Seebeck effect were measured by a Quantum Design PPMS with thermal transport option from 5 to 300 K in a magnetic field up to 9 T.

Normal incidence reflectivity measurements were performed at the SISSI infrared beamline of the ELETTRA Storage Ring (Trieste), between 10 and 30 000 cm\(^{-1}\), at temperatures \(T\) ranging from 5 to 300 K on both samples.

An \textit{in situ} evaporation technique was used to measure the reference. The real part of the optical conductivity \(\sigma(\omega)\) was then determined through Kramers–Kronig (KK) transformations and by standard extrapolations of \(R(\omega)\) both at high and low frequency. Details on the experimental technique and data analysis were reported elsewhere [22].

### 3. Resistivity, Hall effect and Hall mobility

Resistivity measurements for both samples are compared in figure 1(a). The undoped specimen exhibits a pronounced anomaly around \(T \sim 130–140\) K. Then the resistivity decreases monotonically with decreasing temperature. In the inset \(\Delta\rho/dT\) is plotted as a function of \(T\). At \(T = 131.5\) K a cusp is well evident. It has been attributed to the onset of the magnetic transition [7, 9, 23] and in the following is indicated as \(T_{SDW}\). Interestingly, in SmFeAsO the structural transition has been observed at \(130\) K [11], thus suggesting that in this compound the magnetic and structural transitions nearly coincide. This strongly differs from the case of LaFeAsO where the structural transition occurs close to \(160\) K whereas the magnetic one is at \(140\) K. Direct observations of the occurrence of the magnetic transition in SmFeAsO are desirable to clarify this point.

SmFeAs(O0.93F0.07) is characterized by a nearly linear decrease of the resistivity with temperature, with the onset of superconductivity occurring at \(36\) K.

Hall effect measurements are performed at fixed temperature by sweeping the magnetic field from \(-9\) to \(9\) T. A nearly linear dependence of the transverse resistivity on the magnetic field is observed at all the temperatures, different from what is reported for LaFeAsO(\(O_{1−x}F_x\)) [18].

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electron charge). The electron density, $n$, of the two samples is plotted in figure 1(c). The carrier density of the undoped sample strongly depends on temperature, showing a stiff drop corresponding to the resistivity anomaly. This was previously reported in LaFeAsO [9] and was discussed in terms of charge carrier localization at the structural transition. The electron condensation due to the opening of the SDW gap would produce the same feature.

For the 7% F-doped sample, $n$ varies with temperature in a nearly logarithmic way, but no anomaly is observed below $T_{\text{SDW}}$. Interestingly the electron densities of the two samples with increasing temperature above $T_{\text{SDW}}$ tend to overlap and the values nearly coincide at 250 K. Similar results have been obtained on the same compound in [20], on LaFeAsO$_{(1-x)0.5}$ in [18] and on NdFeAsO$_{(1-x)0.5}$ in [17].

The Hall mobility of the two samples evaluated as $\mu_H = R_H/\rho$ is plotted in figure 1(d). In SmFeAsO $\mu_H$ strongly increases by two orders of magnitude below $T_{\text{SDW}}$ and reaches a value of about 200 cm$^2$ V$^{-1}$ s$^{-1}$ at 5 K. In [18] a similar behavior with rather lower values was reported for LaFeAsO. Within an SDW framework, below $T_{\text{SDW}}$ the charge carriers which are gapped out disappear, while the mobility of the carriers which do not condensate in the SDW state abruptly rises. This suggests that electron–electron scattering is the mechanism which mainly limits the carrier mobility above the SDW transition.

The drop of the resistivity below $T_{\text{SDW}}$, despite the strong reduction in carrier density, can be taken into account by an increased mobility of the carriers which do not participate in the SDW state.

The mobility of SmFeAsO$_{(0.93)F_{0.07}}$ decreases with increasing temperature and, also in this case, there is no evidence of the SDW transition. Interestingly, above $T_{\text{SDW}}$, $\mu_H$ in the doped sample is larger than in the undoped one. This follows straightforwardly by the lower resistivity values and the nearly equal carrier density of the former sample in comparison with the latter one. Even if, in multiband conduction, $\mu_H$ is only an effective quantity, these results suggest that doping does not strongly modify the electronic structure; similar results come out from the analysis of the thermal properties [8], which suggests that the density of states is only slightly modified by F doping. Our results do not confirm recent theoretical calculations [24] which predict that doping moves the Fermi level into a region of heavier carriers, rather supporting a scenario in which the Fermi energy is not much shifted by doping [25].

4. Magnetoresistivity

Magnetoresistivity measurements can give further hints into the actual mobility of the two samples. Magnetoresistivity of the doped sample reported in [21] is quite low, indicating low mobility of carriers. It can be roughly estimated of the order of 1% at 50 K and 9 T. Much richer phenomenology is presented by the parent compound, whose magnetoresistivity...
A remarkable positive magnetoresistance is visible at low temperature which progressively disappears with increasing temperature towards the SDW transition, as reported also for LaFeAsO. This is even more evident in figure 3(a) where $\Delta \rho/\rho(0) = \rho(B) - \rho(0)/\rho(0)$, measured at a fixed temperature with increasing magnetic field, is plotted. At 5 K and 9 T $\Delta \rho/\rho(0)$ is more than 15% and with increasing temperature decreases down to 1% at 120 K. $\Delta \rho/\rho(0)$ measured at higher selected temperatures (140, 160, 180, 240 and 300 K) suddenly drops down to 0.1%.

We can further notice that the application of the magnetic field does not affect the SDW transition at all. As shown in the inset of figure 2 $d\rho/dT$, which underlines the SDW, does not present any shift and/or enlargement due to the application of the magnetic field.

The temperature dependence of magnetoresistance can be accounted for by Kohler’s rule which assumes $\Delta \rho/\rho(0)$ as a function of the product $B\tau$, where $\tau$ is the relaxation time: $\Delta \rho/\rho(0) = f(B\tau)$. The relaxation time $\tau$ is related to the resistivity by $\rho = m^*/ne^2\tau$, where $m^*$ is the effective mass. If the factor $m^*/ne^2$ does not change with temperature Kohler’s rule can be written in the more common form $\Delta \rho/\rho(0) = f(B/\rho(0))$. For SmFeAsO the electron density $n$ is far too constant below the SDW, rather it varies within two orders of magnitude as shown in figure 1(b). This avoids the usual scaling $\Delta \rho/\rho(0)$ versus $B/\rho$ working, as is evident in figure 3(b), and led us to conclude that Kohler’s rule is violated. In figure 3(c), $\Delta \rho/\rho(0)$ is plotted as a function of $BR_H/\rho(0) = B\mu_H = B\tau/m^*e = \omega_c\tau$, where $\omega_c$ is the cyclotron frequency. In this case all the curves collapse together showing that the application of Kohler’s rule in a more general way captures the main features of magnetoresistance in a quite extended range of $\omega_c\tau(0–0.16)$. Small differences between the curves, within ten per cent overall, can be appreciated looking at figure 3(c). Such deviations can be attributed to the presence of more bands that participate in the conduction. Moreover, with varying temperature, scattering mechanisms which contribute differently to magnetoresistance, can come into play. For instance, it was emphasized that below 20 K the resistivity is affected by the ordering of the Sm$^{3+}$ sublattice; in such a case the magnetic field might suppress spin fluctuation, giving a negative contribution to magnetoresistance, which sums with the positive cyclotronic contribution.

5. Seebeck effect

The Seebeck coefficient, $S$, of SmFeAsO and SmFeAs(O$_{0.93}$F$_{0.07}$) is shown in figure 4.
Figure 5. Far-infrared reflectivity (a) and optical conductivity (b) at selected temperatures for the undoped SmOFeAs sample. The inset of (a) shows the reflectivity at 300 K between 50 and 20000 cm$^{-1}$ and that of (b) the difference $\Delta \sigma_1 = \sigma_1(\omega, T) - \sigma_1(\omega, 150 \text{ K})$. (c) and (d): same as (a) and (b), respectively, for the superconducting SmFeAs(O$_{0.93}$F$_{0.07}$) sample. In the inset the ratio $R(\omega, T)/R(\omega, 42 \text{ K})$ (see text) is also reported.

In both samples $S$ is negative over the entire temperature range, indicating that electrons dominate the electrical conduction. This is consistent with the measured negative Hall coefficient (figure 1(b)).

The two curves present clear signatures of the different electronic transition occurring in the two samples. In the undoped sample, $S$ presents a decrease of its absolute value below $T_{\text{SDW}}$; similar behavior has been observed in LaFeAsO [9].

Anomalies of this type have been observed below magnetic instabilities [26–28] and can be understood within a free-electron model where $S$ is given by

$$S(T) = -\frac{\pi^2 k_B}{3 |e|} k_B T \left[ \frac{N(E_F)}{n} + \frac{1}{\tau} \frac{d \tau}{d E_F} \right]$$

where $N(E)$ is the density of states, $E_F$ is the Fermi energy and $\tau$ is the relaxation time. The first term scales as $1/E_F$ and does not change abruptly at the transition, while the second one, which can be qualitatively related to the changes of mobility with doping and temperature (see figure 1(d)), gives the main contribution below the transition. Below $T_{\text{SDW}}$, indeed, $\mu_H$ strongly decreases with electron doping, suggesting that $d \tau/dE_F$ is large and negative. Above the transition $\mu_H$ is roughly constant with doping and the second term becomes negligible.

In SmFeAs(O$_{0.93}$F$_{0.07}$) $S$ drops to zero below 34 K in reasonable correspondence with the resistive superconducting transition. Actually, it does not reach zero, but it shows a tail which extends to the lowest temperature, indicating that a minor amount of the sample does not attain the superconducting state.

Neglecting the anomalies related to the ordering transitions, $|S|$ of the two samples has a similar overall behavior, showing a large maximum around 60–70 K. The values are more than three times higher in the doped than in the undoped samples, in agreement with what is observed in LaFeAsO [9] and in LaFeAs(O$_{0.89}$F$_{0.11}$) [29]. Looking at equation (1) it should be the opposite, since $S$ decreases its absolute values with increasing $E_F$. This can be explained considering the multiband nature of the oxypnictides. For two bands, one electron- and the other hole-like, $S$ becomes

$$S = \frac{\sigma_h |S_h| - \sigma_e |S_e|}{\sigma_h + \sigma_e}$$

where $\sigma_{e(h)}$ and $S_{e(h)}$ are the contributions of electrons (holes) to the electrical conductivity and Seebeck coefficient, respectively. Looking at equation (2), the undoped sample might show smaller $S$ values since large but nearly equal hole and electron contributions compensate for each other, whereas electron doping enhances the electron contribution.

6. Infrared reflectivity

The far-infrared $R(\omega)$ is shown in figures 5(a) and (c) at selected $T$ for SmFeAsO and SmFeAs(O$_{0.93}$F$_{0.07}$).
respectively. \( R(\omega) \) is also reported in the whole spectral range at 300 K in the insets of the same figures. The reflectivity of both samples resembles that of a bad metal, showing a weak \( T \) dependence in the infrared and strong phonon peaks (that will be discussed in a forthcoming paper [30]) at approximately 102, 260, 270, 375 and 450 cm\(^{-1}\). Calculations [30] show that the peaks observed at 102, 260 and 450 cm\(^{-1}\) correspond to optical phonons polarized along the \( c \) axis, while those at 270 and 375 cm\(^{-1}\) (which splits into two components at low \( T \)) are in the \( ab \) plane. The presence of both polarizations is in agreement with the polycrystalline nature of the samples. The comparison of our data with some optical \( ab \)-plane results recently appeared on (Ba\(_{1-x}\)K\(_x\))Fe\(_2\)As\(_2\) [31] and LaFePO [32] single crystals and with ellipsometric data obtained on polycrystalline materials [33] shows that the main contribution to the optical properties of pellets comes from the more insulating \( c \) axis, whose optical properties have never been measured to our knowledge in the pnictides.

As discussed previously, the electronic and magnetic properties of the undoped material are affected by an SDW instability of the underlying Fermi surface. This transition determines a strong change in the \( T \) dependence of the resistivity (see figure 1(a)): \( \rho \) is nearly constant for \( T > 135 \text{ K} \), and rapidly decreases at lower \( T \).

The SDW transition affects also the infrared spectrum of the undoped sample, which shows for \( T < T_{SDW} \) (in agreement with similar measurements on LaFeAsO [2]) a suppression of \( R(\omega) \) between 250 and 150 cm\(^{-1}\) and a more pronounced metallic behavior below the lowest-energy phonon absorption (see figure 5(a)). This suppression could not be observed (figure 5(c)) in the doped material. Here, due to the absence of the SDW instability, \( R(\omega) \) is more metal-like and shows a monotonic increase at any \( T \) for \( \omega \to 0 \).

The effect of the SDW transition can be tracked in \( \sigma_1(\omega) \), which is plotted for the undoped and doped sample in figures 5(b) and (d), respectively. At variance with conventional SDW materials like (TMTSF)\(_2\)PF\(_6\) [34], the transition does not open a gap in the electronic excitations in agreement with transport results (see above) but it induces just a small depression in \( \sigma_1(\omega) \) between 250 and 150 cm\(^{-1}\). This depression can be better observed in the inset of figure 5(b) where the difference \( \Delta \sigma_1(\omega) = \sigma_1(\omega, T) - \sigma_1(\omega, 150 \text{ K}) \) is shown at selected \( T < 150 \text{ K} \) and corresponds to a transfer of spectral weight (SW) from high frequencies to those below 100 cm\(^{-1}\). The agreement between the dc results (see above) and the IR data across \( T_{SDW} \) suggests that the SDW transition partially gaps the Fermi surface. Therefore, the metallic term observed at very low frequency is due to the ungapped states around \( E_F \). Its increase below \( T_{SDW} \) can be associated with a reduction of the electron–electron scattering.

In the superconducting SmFeAsO\(_{0.93}F_{0.07}\) \( \sigma_1(\omega) \) is still strongly influenced by the \( c \)-axis conductivity showing phonon absorptions similar to those observed in the undoped material. However, \( \sigma_1(\omega) \) does not show any imprint of the SDW transition, in agreement with the linear decrease of the resistivity and with the other transport data (see above). It monotonically increases at any \( T \) below 600 cm\(^{-1}\).

The effect of the superconducting transition can be directly observed in the inset of figure 5(d), where the ratio \( R(\omega, T)/R(\omega, 40 \text{ K}) \) is reported at different \( T \). Therein, \( R(\omega, T) \) is the reflectivity in the superconducting state at selected \( T < T_c \) and \( R(\omega, 40 \text{ K}) \) is that of the normal state. The well-evident maximum around 30 cm\(^{-1}\) in agreement with similar data collected on polycrystalline (Nd,Sm)FeAs(O\(_{0.82}\)F\(_{0.18}\)) [33], can be associated with the \( c \)-axis superconducting (SC) response. Indeed, if in the normal state the \( c \) axis is nearly semiconducting, below \( T_c \) it becomes metallic due to the Josephson interplane coupling. Therefore the low-frequency \( R(\omega, T)/R(\omega, 40 \text{ K}) \) allows one to evaluate the \( c \)-axis Josephson plasma frequency, which turns out to be about 30 cm\(^{-1}\).

At about 100 cm\(^{-1}\), \( R(\omega, T)/R(\omega, 40 \text{ K}) \) becomes close to 1 as the \( c \)-axis phonon in this spectral region is practically independent of \( T \). Therefore the two deep minima centered around 75 and 125 cm\(^{-1}\) can be interpreted in terms of a single broad minimum with an onset at about 200 cm\(^{-1}\). Previous studies on cuprates have shown that this onset approximately indicates the maximum value of the \( ab \)-plane SC optical gap. With \( 2\Delta \approx 200 \text{ cm}^{-1} \) one has \( 2\Delta/k_B T_c \approx 8 \), much larger than the BCS value of 3.53 which holds for weak coupling. Comparable gap amplitudes are reported for the high-\( T_c \) cuprates, where they point toward a strong-coupling SC pairing mechanism. However recent photoemission data on (Ba\(_{1-x}\)K\(_x\))Fe\(_2\)As\(_2\) [35] have shown that two different gaps open below \( T_c \). The largest one (\( \Delta \approx 100 \text{ cm}^{-1} \)) agrees with the present IR data, while the smallest one (\( \Delta \approx 50 \text{ cm}^{-1} \)) has not been observed up to now by optical measurements. In the latter case one would have \( 2\Delta/k_B T_c \approx 4 \), in much better agreement with other experimental techniques.

7. Conclusions

We report measurements of several transport properties (resistivity, magnetoresistivity, Hall effect, Seebeck coefficient, IR reflectivity) of undoped SmFeAsO and lightly doped SmFeAs(O\(_{0.93}\)F\(_{0.07}\)) oxypnictides. The main purpose was to investigate the effect of the SDW transition on their transport properties and to look for some memory of it at low levels of doping.

All the properties measured on SmFeAsO show clear signatures of the magnetic instability. Resistivity measurements show a maximum at about 135 K followed by a sharp drop below 130 K. Hall effect measurements demonstrate that below this temperature the number of carriers abruptly decreases as a consequence of carrier condensation due to the opening of an SDW gap at the Fermi level. Within this framework, the reduction in the resistivity can be explained with an increased mobility of the carriers which remain free, suggesting a strong electron–electron scattering. The presence of high-mobility carriers in the SDW ordered state is fully supported by the measured magnetoresistivity which is rather large at low temperatures and drastically reduces with increasing temperatures up to \( T_{SDW} \). This behavior is consistent with the Kohler rule, only if the temperature dependence of the carrier density is considered.

Also the Seebeck coefficient of SmFeAsO exhibits a clear signature of the SDW transition: below \( T_{SDW} \), \( S \) changes slope,
indicating that a contribution with opposite sign adds to the main term. This additional term arises by the abrupt change of the carrier scattering rate below $T_{SDW}$. Finally, the effect of the SDW transition can be tracked in the IR response. At variance with conventional SDW materials, the ordering transition does not open a gap in the electronic excitations. It induces just a small depression in $\sigma(\omega)$ between 250 and 150 cm$^{-1}$, suggesting that the transition partially gaps the Fermi surface. Therefore, the metallic term observed at very low frequency is due to the ungapped states around $E_F$. Therefore, the metallic term observed at very low frequency is due to the ungapped states around $E_F$ and its increase below $T_{SDW}$ can be associated with a reduction of the electron–electron scattering.

Concerning the doped sample, transport properties of SmFeAs(O$_{0.89}$F$_{0.07}$) present a more metallic behavior consistent with electron doping, even if the room-temperature values of all the considered quantities nearly overlap in the two samples. This result indicates that F doping does not produce substantial changes in the density of states, nor in the effective mass, as suggested also by thermal properties [8]. However, with decreasing temperature the rich and self-consistent phenomenology summarized up to now completely disappears: all the aforementioned anomalies are missing, in favor of the occurrence of superconducting transition at around 34 K. Here, the effect of this latter has also been directly observed in the far-infrared reflectivity. This allowed us to evaluate the superconducting gap of SmFeAs(O$_{0.89}$F$_{0.07}$), which points towards a pairing mechanism governed by a strong-coupling regime.

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