Intrinsic charge dynamics in high-$T_c$ AFeAs(O,F) superconductors

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We report the first determination of the in-plane complex optical conductivity of 1111 high-$T_c$ superconducting iron oxyxnitride single crystals PrFeAsO0.6F0.35 and thin films SmFeAsO1−xFx by means of bulk-sensitive conventional and micro-focused infrared spectroscopy, ellipsometry, and time-domain THz transmission spectroscopy. A strong itinerant contribution is found to exhibit a dramatic difference in coherence between the crystal and the film. Using extensive temperature-dependent measurements of THz transmission we identify a previously undetected $2.5$-meV collective mode in the optical conductivity of SmFeAs(O,F), which is strongly suppressed at $T_c$ and experiences an anomalous $T$-linear softening and narrowing below $T^* ≈ 110$ K $> T_c$. The suppression of the similar gap with a similar gap ratio $2\Delta/k_B T_c$ $≈ \sim 7$ in both materials, indicating strong pairing.

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Almost a decade of intensive research into the phenomenology of high-transition-temperature (high-$T_c$) iron-based superconductors has revealed that the $T_c$ in the vast majority of these compounds is limited to below about 40 K (Ref. 1). Two notable exceptions to this rule are the oxynitrides of the 1111-type AFeAs(O,F) family (A stands for a rare-earth metal) with $T_c$’s up to about 55 K (Ref. 3) and the monolayer FeSe grown on SrTiO3 (Ref 4, 7) with a $T_c \approx 65$ K. It is now clear, that in the latter case the normally high $T_c$ is afforded not only by the electronic structure and interactions inherent in the iron pnictides and chalcogenides but also by additional, extrinsic, interfacial interactions of itinerant carriers in FeSe with the SrTiO3 substrate (Ref 13-15). In the absence of the latter, the maximum $T_c$ attainable in monolayer FeSe was found to only reach the aforementioned limit of $\sim 40$ K (Ref 6, 14, 17).

These experimental observations emphasize the uniqueness of the superconducting state in the AFeAs(O,F) materials as they reach the 55-K mark unassisted by extrinsic interactions and hold the key to our understanding of the mechanism of high-$T_c$ superconductivity and further enhancing the superconducting transition temperatures in iron-based compounds. Unfortunately, high-quality single crystals of these materials can only be obtained by a laborious high-pressure growth technique (Ref 18), which produces microscopic samples. Small linear dimensions and mass effectively bar a large number of bulk-sensitive experimental techniques from contributing to our knowledge base of iron-oxynitride phenomenology. After several pioneering optical works on polycrystalline 1111-type samples at the dawn of the iron-pnictide research (Ref 19-21), few further attempts have been made at determining the intrinsic optical itinerant response of iron oxynitrides within the superconducting FeAs-layers and its modification in the superconducting state (Ref 22, 23). Another major complication is the polar character of the cleaved crystal planes, which leads to excess charge on the sample surface and makes the interpretation of angle-resolved photoemission spectroscopy as well as scanning-tunneling spectroscopy measurements far from trivial (Ref 24-27). Currently no consensus exists regarding the bulk electronic structure of iron oxynitrides. It is, therefore, imperative to investigate the charge dynamics of these materials and its modification in the superconducting state using

![Figure 1](image_url)

**FIG. 1:** (a) Temperature dependence of the ac magnetic susceptibility of PrFeAsO0.6F0.35 cooled in a zero (blue line) and a 10 Oe (black line) magnetic field, subsequently measured in a 10 Oe field in both cases. The deviation from perfect diamagnetism $\chi = -1$ is due to the geometric factor. (inset) Mass and dimensions of the sample used in the present optical study. (b) Temperature dependence of the dc resistivity of the optimally doped SmFeAs(O,F) thin film. (inset) Schematic illustration of the sample geometry and the doping mechanism by fluorine diffusion upon annealing.
a bulk-sensitive probe of the electronic structure and interactions.

In this Letter, we report the results of a bulk-sensitive broadband optical-spectroscopy study that overcomes the aforementioned materials-related challenges. We use two complementary approaches to shed first direct light onto the bulk charge-carrier response of iron oxypnictides and its modification in the superconducting state. The first approach employs conventional and micro-focused Fourier-transform infrared reflectance spectroscopy as well as micro-focused CCD-based spectroscopic ellipsometry to investigate the intrinsic electrodynamics of microscopic high-pressure-grown \[\text{PrFeAsO}_{0.6}\text{F}_{0.35}\] single crystals of \text{PrFeAsO}_{0.6}\text{F}_{0.35} (see Fig. 2a) in a wide spectral range from 15 meV to 6 eV. The second approach makes use of a unique fluorine-diffusion doping process by means of \textit{in situ} annealing after growth of non-superconducting parent \text{SmFeAsO} single-crystalline thin films synthesized by state-of-the-art molecular beam epitaxy on a CaF\[2\] substrate and capped by a SmF\[3\] layer \[28\] (see Fig. 2h). This approach has been shown to result in high-quality optimally doped iron-oxypnictide \text{SmFeAs(O,F)} thin films with a maximum \(T_c = 55\) K. We have carried out extensive synchrotron- and thermal-source-based variable-angle-of-incidence spectroscopic ellipsometry as well as time-domain THz transmission spectroscopy measurements on these films in the range from 1 meV to 6.5 eV and at temperatures from 4 to 300 K. These comprehensive measurements allowed us to extract the complex optical conductivity of the \text{SmFeAs(O,F)} thin film from that of the total response of the multilayer structure and access the itinerant-carrier dynamics in this material down to energies well below those afforded by the linear dimension of single-crystalline microcrystals.

The central observations of our work are summarized in Fig. 2. We find high values of the infrared reflectance in the \text{PrFeAsO}_{0.6}\text{F}_{0.35} microcrystal (Fig. 2a), indicative of a strong itinerant response. By means of a Drude-Lorentz fit \[29\] we extract the total plasma frequency of 1.4 eV — on par with many other iron-based superconductors \[10\] despite the extremely singular band structure of the 1111-type materials \[25, 27\]. This itinerant response reaches a very high degree of coherence at lowest temperatures (quasiparticle scattering rate \(\gamma\) of about 5 meV) — a property of materials with low levels of crystalline disorder. Below \(T_c = 24\) K the infrared reflectance approaches unity below the characteristic energy \(E_0 = 28\) meV indicative of the opening of a nodeless superconducting gap \[30, 31\]. Such a high gapping energy is remarkable for a superconductor with \(k_B T_c \approx 2\) meV.

In the \text{SmFeAsO}_{1-x}\text{F}_x thin film, analogous Drude-Lorentz decomposition of the optical conductivity (shaded areas in Fig. 2b) reveals an equally strong itinerant response but significantly less coherent (as can also be seen from the direct comparison with the optical conductivity of the \text{PrFeAsO}_{0.6}\text{F}_{0.35} microcrystal shown as a dotted line in Fig. 2a). The quasiparticle scattering rate is found to be 150 meV at 300 K and remains unchanged down to lowest temperatures, thus exceeding its \text{PrFeAsO}_{0.6}\text{F}_{0.35} counterpart by almost two orders of magnitude.

By virtue of the large surface area of our \text{SmFeAsO}_{1-x}\text{F}_x thin film we were able to investigate its optical response deep in the infrared regime extending to sub-THz frequencies. This unprecedented for the 1111-type iron oxypnictides spectroscopic access uncovered the existence of a low-energy collective mode, manifested as a broad peak in the optical conductivity centered at 2.5 meV at room temperature. The peak narrows and grows dramatically with decreasing temperature, approaching 15 m\(\Omega\)\(^{-1}\)cm\(^{-1}\) at its maximum — an order of magnitude larger than the normal-state dc resistivity values of up to 2 m\(\Omega\)\(^{-1}\)cm\(^{-1}\). It then rapidly decreases at lower photon energies to values consistent with the dc transport. The detection of this collective mode requires robust optical access to energies below 3 meV and has not been observed previously in any iron pnictide or chalcogenide.

In the superconducting state, we find a strong signature of a coherent superconducting condensate, manifested in the dors-
The low-energy collective mode shows dramatic sensitivity to both \( T_c \) and \( T^* \). To demonstrate it, we fit the energy dependence of the real part of the THz conductivity in the THz spectral range (Fig. 2b). The real part of the optical conductivity in Fig. 2b is likewise sensitive to the onset of superconductivity and allows us to extract the superconducting energy gap in what follows. Finally, we discover a distinct temperature scale of \( T^* = 110 \text{ K} \gg T_c \) (black arrow in Fig. 2b), at which the real part of the optical conductivity displays an additional thermal anomaly.

We hypothesize that the observed collective mode could originate in the quantum critical fluctuations of incommensurate density-wave order. Density-wave fluctuations/order have been found in both the spin- and, possibly, charge \( [33, 34] \) channel in proximity to the optimally doped iron-oxypnictide superconductors. The hydrodynamic description of these fluctuations indicates that they should manifest themselves as a low-energy collective mode in the optical conductivity of strongly correlated bad metals \([35]\), such as iron-based superconductors \([10]\). Both the mode energy \( \hbar \omega_0 \) and width \( \hbar \Gamma \) are predicted to exhibit a conspicuous linear temperature dependence, \( \hbar \omega_0 \sim \hbar \Gamma \sim k_B T \), analogous to the \( T \)-linear dc resistivity observed in many unconventional superconductors \([27, 38]\). Fig. 3b shows that both \( \hbar \omega_0 \) and \( \hbar \Gamma \) of the collective mode in the SmFeAs(O,F) thin film display a clear linear temperature dependence below \( T^* \), consistent with the aforementioned interpretation.

In both the PrFeAs\(_{0.6}F_{0.35}\) microcrystal and the SmFeAs(O,F) thin film the onset of superconducting coherence is manifested in the transfer of a portion of the infrared spectral weight (hatched areas in Figs. 4a,c) to the dissipationless response at zero frequency according to the Ferrell-Glover-Tinkham sum rule \([39]\). This spectral weight corresponds directly to the London penetration depth of a superconductor and in our analysis amounts to \( \lambda_{\text{L}} = 190 \pm 100 \text{ nm} \) in the microcrystal and a significantly larger \( \lambda_{\text{L}} = 550 \pm 50 \text{ nm} \) in the thin film.

The signatures of the superconducting optical gap are best
revealed in the ratio of the optical conductivity below $T_c$ to that in the normal state just above $T_c$: $\sigma_1(\omega) = \sigma_{1N}(\omega)/\sigma_{1S}(\omega)$. We examine these ratios for the case of the PrFeAsO$_{0.6}$F$_{0.35}$ microcrystal and SmFeAs(O,F) thin film in Figs. 3, 4, respectively. Corresponding to the near-unity reflectance below $E_0$ in the superconducting state of PrFeAsO$_{0.6}$F$_{0.35}$ in Fig. 3, $\sigma_1(\omega)$ for this material vanishes below the same energy. In a conventional superconductor with a high impurity concentration, the onset of absorption in the superconducting state occurs when the photon energy is sufficient to dissociate a Cooper pair with the binding energy of 2$\Delta$ [32, 40]. However, we have demonstrated earlier (see Fig. 2 and the corresponding discussion in the text), that PrFeAsO$_{0.6}$F$_{0.35}$ microcrystals exhibit a high degree of coherence at low temperatures. In such a clean superconductor, the direct dissociation of a Cooper pair by an incident photon in a two-body process is forbidden by the conservation of energy and momentum. For optical absorption to occur at low temperatures, a quantum of the field mediating the pairing interaction must be excited in addition. If the excitation spectrum of the mediating boson is gapped up to the energy $E_g$ then absorption becomes allowed above 2$\Delta + E_g$ (Ref. 41). In iron-based superconductors the mediating interaction is believed to be of spin-fluctuation origin and indeed has a gapped excitation spectrum in the superconducting state [2], with the spin-gap energy $E_g$ reaching 2$\Delta$ [42, 43]. The combination of multiple Andreev reflection spectroscopy [44] and powder inelastic neutron scattering [45] clearly demonstrate that the gap energy in the family of 1111-type materials is $E_g \approx 2\Delta$. Therefore, optical absorption in the superconducting state is expected to occur at an energy of 2$\Delta + E_g \approx 4\Delta$, which in the present case results in $\Delta \approx 7$ meV and a gap ratio of 2$\Delta/k_BT_c \approx 7$, in a good agreement with the largest values found in the pnictides in general [2] and, more importantly, in the materials of the same family via ARPES [26]. Signatures of mediating-boson-assisted absorption in the infrared conductivity have been previously identified in Ba$_{1-x}$K$_x$Fe$_2$As$_2$, BaFe$_2$(As$_{0.67}$P$_{0.33}$)$_2$, and NaFeO$_{0.978}$CoO$_{0.022}$As in Refs. [29, 43, 46] respectively.

Similarly to the case of PrFeAsO$_{0.6}$F$_{0.35}$, $\sigma_1(\omega)$ below $T_c$ in the optimally doped SmFeAs(O,F) thin film reveals a plateau below an energy of about 33 meV (see Fig. 2), albeit the absorption does not vanish completely at any photon energy and a sizable residual optical conductivity is present (this observation is consistent with the previous steady-state and ultrafast spectroscopy measurements on 1111-type single crystals and thin films [22, 23]). In this case, the significantly less coherent itinerant response than in the PrFeAsO$_{0.6}$F$_{0.35}$ microcrystal allows for a direct dissociation of the Cooper pairs without the assistance of the mediating boson, as the excess momentum is taken up by the lattice via defects. One may thus expect that the standard Mattis-Bardeen expression for the anomalous skin effect in an impure superconductor with a nodeless gap [32] should apply. Indeed, we find that our experimental data are very well reproduced by this theory (grey line in Fig. 4). The nodeless character of the superconducting gap is consistent with previous studies of 1111-type compounds [23, 26, 27, 44, 47]. The observed agreement between experiment and theory allows us to assign the energy of 33 meV directly to the binding energy of the Cooper pair, 2$\Delta$, which results in a gap ratio 2$\Delta/k_BT_c \approx 7.2$. This value is remarkably similar to that in PrFeAsO$_{0.6}$F$_{0.35}$ and, furthermore, to the largest gap ratio identified via ARPES in NdFeAsO$_{1-\delta}$F$_x$ (Ref. 26) and optimally doped Ba$_{0.68}$K$_{0.32}$Fe$_2$As$_2$ (Refs. 48, 49). This commonality suggests a single pairing mechanism in all of these compounds and a strong coupling between electrons and the pairing boson. Our work paves the way to future systematic spectroscopic studies of the in-plane infrared charge response of the high-Tc 1111-type iron oxypnictides. Such investigations will enable the extraction of the spectral function of the pairing boson [29, 43, 50] and its evolution across the phase diagram, shedding light onto the microscopic origin of the highest bulk superconducting transition temperature among the iron-based superconductors.

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