Interlayer Coherence and Superconducting Condensate in the c-Axis Response of Optimally Doped $Ba(Fe_{1-x}Co_x)_2As_2$ High-T_c Superconductor Using Infrared Spectroscopy

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We report on the infrared studies of the interlayer charge dynamics of a prototypical pnictide superconductor $Ba(Fe_{0.926}Co_{0.074})_2As_2$. We succeeded in probing the intrinsic interlayer response by performing infrared experiments on the crystals with a cleaved *ac* surface. Our experiments identify the coexistence of the suppression of the electronic spectral weight and the development of a coherent Drude-like response in the normal state. The formation of the interlayer condensate is clearly observed in the superconducting state and appears to be linked to coherent contribution to the normal-state conductivity.

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Superconductivity at a genuinely high-transition temperature T_c exceeding 40–50 K is only observed in layered materials such as cuprates and iron-based compounds. Unraveling the role of electronic anisotropy imposed by the layered crystal structure constitutes an important step toward the understanding of the high-T_c phenomenon. The degree of the electronic anisotropy in the cuprates can vastly vary; recent studies revealed that superconductivity can still occur when two-dimensional CuO_2 planes are essentially decoupled [1–4]. The iron-based superconductors (Fe-SCs) are known to be more isotropic than the cuprates [5,6] but data on their interlayer charge dynamics are very limited.

Infrared spectroscopy is ideally suited for investigating the anisotropic electrodynamics of layered superconductors. Indeed, infrared studies of the interlayer electrodynamics of the underdoped cuprates have identified two interrelated aspects of the phenomenology of these materials: the normal-state pseudogap and condensate formation from incoherent conductivity [3,7]. The interlayer conductivity $\sigma_{1c}(\omega)$ in the normal state is characterized as incoherent in view of the absence of any well-defined Drude peak. The formation of an interlayer condensate occurs through further suppression of this incoherent response over a broad frequency region dramatically exceeding the superconducting energy gap.

Current experimental information on the interlayer electrodynamics of the Fe-SCs is incomplete and riddled with inconsistencies. Most of the infrared studies of the Fe-SCs are focused on the *ab*-plane response [8–17] and only two works explored the c-axis response of the superconducting compounds, optimally doped (OPD) $Ba_{0.67}K_{0.33}Fe_2As_2$ (Ref. [18]) and $FeTe_{0.55}Se_{0.45}$ (Ref. [19]). Unexpectedly, the c-axis optical spectra in Refs. [18,19] showed virtually no changes below T_c, suggesting that the interlayer component of the superfluid density tensor $\rho_{s,c}$ is vanishingly small. This finding implies a much stronger anisotropy of the superconducting state compared to what is registered in other probes [6]. Furthermore, this result led to a proposal that the superconducting gap of OPD $Ba_{1-x}K_xFe_2As_2$ has nodes at the portion of Fermi surfaces dominating the c-axis conduction [18]: a conjecture that is in contradiction with the results of recent thermal conductivity [20] and angle-resolved photoemission measurements [21] on OPD $Ba_{1-x}K_xFe_2As_2$. Generally, the c-axis condensate can be readily detected with infrared methods [3] and only a very limited subset of the cuprates where superconductivity coexists with spin and/or charge order reveals drastic suppression of $\rho_{s,c}$ below the measurements limits [4,22]. We note that the spin or charge order might also be expected in the Fe-SCs [23,24]. Before drawing any further inferences from the oddities of available c-axis data for the Fe-SCs, it is important to realize that the above infrared experiments have been carried out for single crystals with polished *ac* surface [18,19]: a procedure that may introduce artifacts in the optical data possibly due to surface damage and/or stain as exemplified in Refs. [25–28]. Intrinsic c-axis properties that can only be obtained for as-grown or cleaved surfaces are still not available thus hindering progress with the understanding of the electronic anisotropy of the Fe-SCs.

In this Letter, we report on the first infrared studies of the interlayer charge dynamics of a prototypical pnictide superconductor $Ba(Fe_{0.926}Co_{0.074})_2As_2$ (BaCo122) at doping level yielding maximum T_c in this particular phase. We have succeeded in probing the intrinsic c-axis electronic response of OPD BaCo122 superconductor by performing infrared experiments on the crystals with a cleaved *ac* surface. These results unambiguously reveal the robust interlayer superfluid with $\rho_{s,c} \approx (2.3 \pm 0.2) \times 10^6\ \mathrm{cm}^{-2}$ corresponding to the c-axis magnetic penetration depth $\lambda_c = 1050$ nm. The normal-state response of these samples shows gradual depression of far-infrared conductivity with decreasing temperature (T). The spectral weight (SW) removed from low frequencies is shifted to higher energy. Apart from the overall depression in $\sigma_{1c}(\omega)$, we find a presence of the c-axis coherence at the lowest frequencies in the normal state. A hallmark of the interlayer electrodynamics of BaCo122 system is that coherent contribution is primarily responsible for the formation of the interlayer condensate below T_c. Robust interlayer condensate at $T < T_c$ observed in our contactless studies has direct bearing for technical superconductivity demanding the ability to sustain high magnetic field and/or critical currents [28].

Single crystals of OPD $\mathrm{Ba(Fe_{0.926}Co_{0.074})_2As_2}$ and $\mathrm{Ba(Fe_{0.92}Co_{0.08})_2As_2}$ were grown using self-flux method [29,30]. The c-axis reflectance $R_c(\omega)$ of OPD $\mathrm{Ba(Fe_{0.926}Co_{0.074})_2As_2}$ crystal with the size of $1 \times 0.6\ \mathrm{mm}^2$ was measured between 40 and 25000 cm^{-1} using *in situ* overcoating technique [31]. The experimental uncertainty in raw $R_c(\omega)$ data is about 1% in far-infrared frequency region. Details of infrared spectroscopy experiments are given in Supplemental Material [32]. For comparison we measured the ab-plane reflectance $R_{ab}(\omega)$ of OPD $\mathrm{Ba(Fe_{0.92}Co_{0.08})_2As_2}$. We note that the ab-plane response does not change significantly with the variation in Co concentration near optimal doping [9–15,17]. The complex optical conductivity was determined using Kramers-Kronig analysis [33].

Figure 1 displays c-axis reflectance $R_c(\omega)$ of OPD BaCo122 at several T. For comparison, we also plot

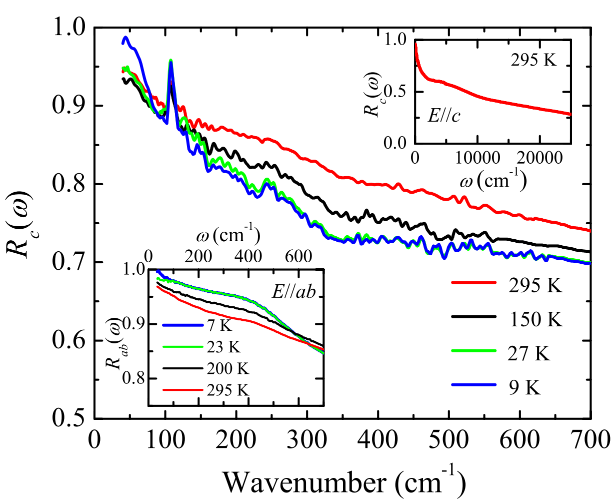

FIG. 1 (color online). T-dependent c-axis reflectance spectra $R_c(\omega)$ of $\mathrm{Ba(Fe_{0.926}Co_{0.074})_2As_2}$ with $T_c = 23$ K. The sharp spike at 108 cm^{-1} is due to infrared active phonon mode. Upper inset: $R_c(\omega)$ over broad frequency up to 25000 cm^{-1}. Lower inset: the ab-plane reflectance $R_{ab}(\omega)$ of OPD $\mathrm{Ba(Fe_{0.92}Co_{0.08})_2As_2}$ in the far-infrared region.

$R_{ab}(\omega)$ spectra of OPD BaCo122 in the lower inset [17]. The ab-plane response is metallic: $R_{ab}(\omega)$ increases at low ω and T. A similar trend is also seen in the $R_c(\omega)$ spectra. However, the T dependence of $R_c(\omega)$ is nonmonotonic. As T is reduced from 295 to 150 K, $R_c(\omega)$ is suppressed in the entire far-infrared region. Below 150 K, $R_c(\omega)$ at $\omega > 100\ \mathrm{cm}^{-1}$ decreases further; below 100 cm^{-1} we observe small increase of $R_c(\omega)$ signaling a crossover to more coherent interlayer dynamics. This T dependence of $R_c(\omega)$ is fully consistent with the interlayer dc transport [5]. Below $T_c = 23$ K, we find an unmistakable infrared signature of the superconductivity [3]: an upturn in $R_c(\omega)$ at about 90 cm^{-1}. We stress that such a change is not observed in the c-axis spectra of OPD BaCo122 with polished ac surface; spectra obtained for the polished samples are shown in Supplemental Material [32]. Moreover, the sharp phonon feature at 108 cm^{-1} in the cleaved crystal becomes drastically suppressed in the polished sample. The same tendencies were registered in the optical response of $\mathrm{Ba_{1-x}K_xFe_2As_2}$ [18]. These results therefore suggest that mechanical polishing is detrimental to probing the intrinsic interlayer behavior of the Fe-SCs.

Insights into the interlayer electronic response can be gained from the analysis of the conductivity spectra. Figures 2(a) and 2(b) show the real part of the c-axis and ab-plane optical conductivity [$\sigma_{1c}(\omega)$ and $\sigma_{1ab}(\omega)$] of OPD BaCo122, respectively. Metallic behavior is evident in the ab-plane response. The Drude-like peak at $\omega = 0$ is clearly observed in $\sigma_{1ab}(\omega)$ data in the normal state. As T decreases, the Drude-like peak becomes narrower and $\sigma_{1ab}(\omega)$ in the low-frequency region is enhanced. In contrast, $\sigma_{1c}(\omega)$ shows a flat spectral form and is gradually suppressed with decreasing T. We will discuss the depletion of the overall level of $\sigma_{1c}(\omega)$ later in detail. Aside from the depression, we find a conspicuous presence of coherence in the interlayer response: a weak metallic upturn in $\sigma_{1c}(\omega)$ below 60 cm^{-1}. The coherence is most evident at $T \approx T_c$: the overall SW in the far-infrared region is depressed compared to data at higher temperatures, whereas the dc conductivity is enhanced. At the lowest T and ω, the remaining conductivity narrows and evolves into a weak Drude-like peak, as can be identified by the rise of $\sigma_{1c}(\omega)$ at the lowest frequencies continued in the region extrapolated to the dc value at 27 K.

The onset of superconductivity is clearly observed in our $\sigma_{1c}(\omega)$ data [Fig. 2(a)]. In the superconducting state $\sigma_{1c}(\omega)$ exhibits substantial depression below about 250 cm^{-1} with SW transfer into the condensate δ peak [3] at $\omega = 0$, consistent with the small residual thermal conductivity of OPD BaCo122 below T_c (Ref. [34]). The Ferrell-Glover-Tinkham (FGT) sum rule states that the missing SW defines the magnitude of the superfluid density as

$$\rho_{s,c} = \frac{c^2}{\lambda_c^2} = \int_0^W [\sigma_{1c}(\omega, T = T_c) - \sigma_{1c}(\omega, T \ll T_c)] d\omega,$$

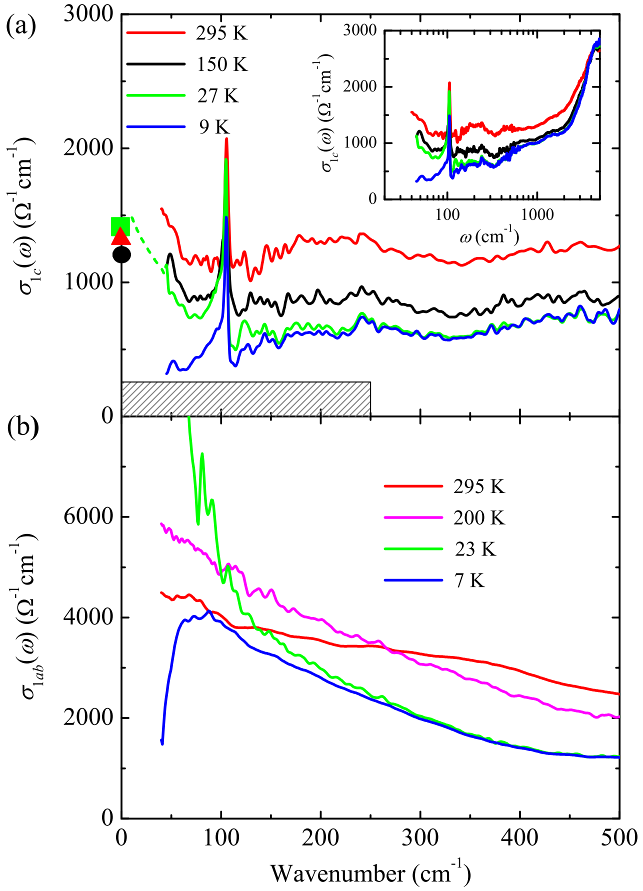

FIG. 2 (color online). (a) Real part of the c-axis optical conductivity $\sigma_{1c}(\omega)$. The sharp peak at 108 cm^{-1} is due to infrared active phonon mode. As T decreases, $\sigma_{1c}(\omega)$ in the far-infrared frequency region becomes depressed and the suppressed SW is most likely not recovered up to 4000 cm^{-1}, as shown in the inset. The dashed line represents the extrapolated $\sigma_{1c}(\omega)$ at $T = 27$ K using the Hagen-Rubens analysis of $R_c(\omega)$. The hatched area denotes the SW of the superconducting condensate. The solid circle, triangle, and rectangle denote the dc conductivity values at 295, 150, and 27 K, respectively (Ref. [5]). (b) Real part of the ab-plane optical conductivity $\sigma_{1ab}(\omega)$ of OPD $Ba(Fe_{0.92}Co_{0.08})_2As_2$ (Ref. [17]).

where λ_c is the c-axis magnetic penetration depth and c is the speed of light [3]. The integral cutoff of $W = 300$ cm^{-1} is sufficient to accommodate the entire region where the spectra undergo changes below T_c. The FGT analysis using a low-frequency extrapolation of $\sigma_{1c}(\omega)$ at $T = 27$ K yields $\rho_{s,c} \approx (2.6 \pm 0.4) \times 10^6$ cm^{-2} ($\lambda_c = 990$ nm). We verified that some ambiguity inevitably introduced by the extrapolation procedure produces only minor variations of $\rho_{s,c}$ [32]. This analysis along with the form of $\sigma_{1c}(\omega)$ data shows that most of the condensate originates from the suppression of the low-frequency Drude-like response at 27 K.

Our assertion of the formation of a δ peak is supported by our analysis of the imaginary part of the conductivity $\sigma_{2c}(\omega)$. The magnitude of $\rho_{s,c}$ can be obtained from

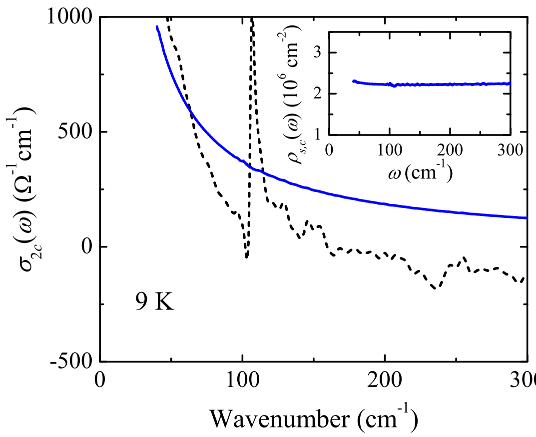

FIG. 3 (color online). Imaginary part of c-axis optical conductivity $\sigma_{2c}(\omega)$ of $Ba(Fe_{0.926}Co_{0.074})_2As_2$ at 9 K. Dashed line: raw $\sigma_{2c}(\omega)$ data. Solid line: $\sigma_{2c}(\omega)$ after the correction for a finite regular contribution characterizing the screening effects that are not related to superconducting carriers. Inset: the c-axis superfluid density $\rho_{s,c}(\omega) = 4\pi\omega\sigma_{2c}(\omega)$.

$\sigma_{2c}(\omega)$: by the Kramers-Kronig relation, the δ peak at $\omega = 0$ in $\sigma_{1c}(\omega)$ implies that $\sigma_{2c}(\omega)$ has the form $\sigma_{2c}(\omega) = \rho_{s,c}/(4\pi\omega)$. However, the latter $1/\omega$ power law is only valid when there is no residual conductivity at $T < T_c$. The raw $\sigma_{2c}(\omega)$ of OPD BaCo122 (dashed line) does not follow the $1/\omega$ dependence and is negative at 160 cm^{-1} (Fig. 3). We found that the deviations from the $1/\omega$ law are caused by the finite values of $\sigma_{1c}(\omega)$ at $T < T_c$. After correcting for this residual contribution according to the standard procedure described in Ref. [35], the $1/\omega$ law in $\sigma_{2c}(\omega)$ is evident over the extended frequency region. The value of $\rho_{s,c}$ is found to be $(2.3 \pm 0.2) \times 10^6$ cm^{-2} ($\lambda_c = 1050$ nm), which agrees well with that obtained from the above FGT sum rule analysis. With the known values of the ab-plane superfluid density $\rho_{s,ab} \approx 2.2 \times 10^7$ cm^{-2} ($\lambda_{ab} = 340$ nm) from Refs. [9–14] as well as our own data [Fig. 2(b)], we find that the anisotropy of superfluid density $\rho_{s,ab}/\rho_{s,c} \approx 7$–11 in OPD BaCo122 in accord with the tunnel diode resonator measurements [36].

The residual conductivity below the threshold structure at 90 cm^{-1} in our $\sigma_{1c}(\omega, T = 9\ \mathrm{K})$ spectra is intriguing. Searching for the origin of this effect we note that this residual conductivity is fully consistent with the magnitude of the c-axis superfluid density that we obtained. Indeed, an assumption that in a "hypothetical" OPD BaCo122 crystals the entire SW below the threshold structure at 90 cm^{-1} in our $\sigma_{1c}(\omega)$ spectra condenses yields an estimate of the superfluid density as high as $\rho_{s,c} \approx (4.1 \pm 0.4) \times 10^6$ cm^{-2} ($\lambda_c = 788$ nm). If true, this value would imply a significantly reduced anisotropy of the superconducting state response that is inconsistent with the literature [36] and with our own data. Heat transport studies of the OPD BaCo122 crystal suggested that the strength of the interlayer pairing may be weaker than that of the in-plane pairing and the former direction is more susceptible to pair breaking [34]. Angle-resolved photoemission spectroscopic experiments of OPD $Ba_{1-x}K_xFe_2As_2$ also showed that the magnitude of the interlayer pairing energy scale is smaller than that of the in-plane pairing energy scale by a factor of 4 [21]. All these findings are in accord with a substantial SW in $\sigma_{1c}(\omega)$ that appears inapt for condensate formation (Fig. 3).

Inhomogeneity of the superfluid density may be responsible for the pair breaking leading to "filling" the superconducting gap with the residual SW that dominates low-frequency behavior of the $\sigma_{1c}(\omega)$ spectra at $T \ll T_c$. A scanning tunneling microscopy study of BaCo122 reported on the spatial variation in the superconducting gap and thus in the superfluid density [37]. The inhomogeneity might be related to disordered Co substitution. Theoretical analyses of the optical response of inhomogeneous superconductors showed that the inhomogeneity of the superfluid density displaces SW from the δ peak to finite frequencies thus triggering the residual conductivity [38,39]. This inference holds irrespective of the particular symmetry of superconducting order parameter.

Having established superconductivity-induced spectral changes, we now proceed to discuss the overall depression of $\sigma_{1c}(\omega)$ in the normal state. Inspection of $\sigma_{1c}(\omega)$ over a broad frequency region reveals that the SW removed from low frequencies (<3000 cm^{-1}) is shifted to higher energy. The global sum rule implies the conservation of total optical SW. Data in the inset of Fig. 2(a) show that the SW lost in the far-infrared region is likely not recovered up to 4000 cm^{-1}. We can rule out that the SW is directed to the frequency region below the low-frequency cutoff of our experiments. Such a transfer would lead to a substantial increase in the raw $R_c(\omega)$ in the frequency range accessible to us: a hypothesis not supported by our infrared data. We also note that the magnitude of $\sigma_{1c}(\omega)$ at the lowest frequency measured at $T = 295$ K and 150 K is close to the dc conductivity [5].

It is worth pointing out that the evolution of $\sigma_{1ab}(\omega)$ with T is similar to that of $\sigma_{1c}(\omega)$ in OPD BaCo122 [15,17]. Whereas a Drude-like peak becomes enhanced with decreasing T, the total SW below 3000 cm^{-1} is suppressed and is shifted to the higher energies in both $\sigma_{1ab}(\omega)$ and $\sigma_{1c}(\omega)$ [17]. As a side remark we mention that the behavior of $\sigma_{1c}(\omega)$ and $\sigma_{1ab}(\omega)$ of BaCo122 is reminiscent of a pseudogap formation registered in the charge dynamics of numerous correlated electron materials including the underdoped cuprates [40]. Pseudogap describes a partial or incomplete gap in the electronic density of states and involves a gradual depletion of low-frequency SW and its transfer to much higher energies. The large energy scale of the SW redistribution is an indication of a crucial role of many-body interactions in a material [40]. The multiband character of BaCo122 is also likely to play a role in the suppression of the electronic SW. Detailed calculations pointing to a role of multiband effects in the SW redistribution are only available for the ab-plane conductivity [41]. We stress that experimental manifestations of these effects are more pronounced in the interlayer data in view of the suppressed Drude contribution. New observations in Fig. 2 call for an in-depth theoretical analysis of the interlayer charge dynamics of the pnictides.

The infrared experiments reported here resolve gross inconsistencies of the interlayer electrodynamics of the Fe-SCs at $T < T_c$ (Refs. [18,20,21]). A finding of note reported here is that polishing can obscure intrinsic characteristics of the infrared response of the Fe-SCs. The normal-state data highlight the coexistence of metallic and localizing trends in the response of the Fe-SCs: a hallmark of many classes of correlated electron systems including the cuprates [40]. This coexistence is most clearly evident in the $\sigma_{1c}(\omega, T \approx T_c)$ spectra in Fig. 2 where signs of interlayer coherence are found side by side with the incoherent response. The coherent contribution is primarily responsible for the formation of the superconducting condensate that we observed for the first time in the interlayer infrared response of Fe-based materials. This latter aspect of superconductivity of the Fe-based system is in stark contrast to the underdoped cuprates. In the latter materials, Josephson coupling between the CuO_2 planes appears to be capable of mobilizing the incoherent c-axis conductivity to produce a strongly anisotropic superfluid response at $T < T_c$.

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