Interplane Transport and Superfluid Density in Layered Superconductors

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We report on generic trends in the behavior of the interlayer penetration depth $\lambda_c$ of several different classes of quasi two-dimensional superconductors including cuprates, $\text{Sr}_2\text{RuO}_4$, transition metal dichalcogenides and organic materials of the $(\text{BEDT}-\text{TTF})_2X$ - series. Analysis of these trends reveals two distinct patterns in the scaling between the values of $\lambda_c$ and the magnitude of the DC conductivity: one realized in the systems with a Fermi liquid (FL) ground state and the other seen in systems with a marked deviation from the FL response. The latter pattern is found primarily in under-doped cuprates and indicates a dramatic enhancement (factor $\approx 10^2$) of the energy scale $\Omega_C$ associated with the formation of the condensate compared to the data for the FL materials. We discuss implications of these results for the understanding of pairing in high-$T_c$ cuprates.

The formation of the superconducting condensate in elemental metals and their alloys is well understood within the theory of Bardeen, Cooper and Schrieffer (BCS) in terms of a pairing instability in the ensemble of Fermi liquid quasiparticles. Applicability of the FL description to high-$T_c$ cuprate superconductors is challenged by remarkable anomalies found in both the spin- and charge response of these compounds in the normal state $\Omega_C$. Because quasiparticles are not well defined at $T > T_c$ in most cuprates it is natural to inquire into the distinguishing characteristics of the superconducting condensate which appears to be built from entirely different "raw material". Infrared spectroscopy is perfectly suited for the task. Indeed, the analysis of the optical constants in far infrared (IR) unfolds the process of the formation of the condensate $\delta(0)$-peak in the dynamical conductivity and also gives insight into the single-particle excitations in a system both above and below $T_c$.

In this paper we focus on the interplane properties of high-$T_c$ superconductors. We will show that the distinctions in the behavior of the condensate in the FL superconductors and high-$T_c$ cuprates are most radical in the case of the $c$-axis interplane response. The analysis of the generic trends seen in the behavior of the $c$-axis condensate (correlation between the penetration depth $\lambda_c$ and the DC conductivity) allows us to infer the energy scale $\Omega_C$ associated with the development of the superfluid. This energy scale is of the order of the energy gap $2\Delta$ in FL superconductors but may dramatically exceed the gap in systems lacking well defined quasiparticles at $T > T_c$ (primarily in under-doped cuprates). We discuss a connection between the magnitude of $\Omega_C$ and the nature of the normal state response.

Experimentally, the inter-layer $c$-axis penetration depth of a layered superconductor can be measured using a variety of techniques such as microwave absorption, magnetization, vortex imaging or IR spectroscopy. In the IR approach, the magnitude of $\lambda_c$ can be reliably extracted from the imaginary part of the complex conductivity $\sigma_1(\omega) + i\sigma_2(\omega)$ as $1/\lambda^2 = \omega \times \sigma_2(\omega)$. Regardless of the method employed, the penetration depth in several families of cuprates reveals a universal scaling behavior with the magnitude of $\sigma_{DC}(T = T_c)$ (Fig. 1) where the absolute value of $\lambda_c$ is systematically suppressed with the increase of the normal state conductivity. The scaling is obeyed primarily in under-doped cuprates. The deviations from the scaling are also systematic and are most prominent in over-doped phases (red symbols in Fig. 1). Such deviations are a direct consequence of a well-established fact: on the over-doped side of the phase diagram the DC conductivity increases whereas $\lambda_c$ is either unchanged or may show a minor increase.

We find a similar scaling pattern in other classes of layered superconductors, including organic materials, transition metal dichalcogenides and $\text{Sr}_2\text{RuO}_4$ (Fig. 2). While the non-cuprate data set is not nearly as dense, the key trend is analogous to the one found for cuprates. The slope of the $\lambda_c - \sigma_{DC}$ dependence is also close for both cuprates and non-cuprate materials. The principal difference is that the cuprates universal line is shifted down by approximately one order of magnitude in $\lambda_c$. The latter result shows that the superfluid density ($\propto 1/\lambda^2$) is significantly enhanced in under-doped cuprates compared to non-cuprate materials with the same DC conductivity.

Possible origins of the $\lambda_c - \sigma_{DC}$ correlation were recently discussed in the literature. A plausible qualitative account of this effect can be based on the Ferrel-Glover-Tinkham (FGT) sum rule:

$$\frac{c^2}{\lambda^2} = \int_{0+}^{\Omega_C} [\sigma_1^N(\omega) - \sigma_1^{SC}(\omega)]d\omega \quad (1)$$

where $\sigma_1^N(\omega)$ is the normal state conductivity and $\sigma_1^{SC}(\omega)$ is the conductivity due to un-paired carriers in the superconducting state at $\omega > 0$ and $T \ll T_c$. For a dirty limit superconductor $\sigma_1^N(\omega) \approx \sigma_{DC}$, and Eq. 1 can be approximated as:

$$\frac{c^2}{\lambda^2} \approx 2\Delta\sigma_{DC}. \quad (2)$$

Such an approximation is possible because within the BCS model the energy scale $\Omega_C$ from which the condensate is collected is of the order of magnitude of the gap:
\(\Omega_C \approx 2\Delta \approx 3 - 5kT_c\). A connection between \(1/\lambda^2\), \(\sigma_{DC}\), and the energy gap is illustrated in the inset of Fig. 1. In the dirty limit the magnitude of \(\sigma_{DC}\) sets the amount of spectral weight available in the normal state conductivity whereas the magnitude of \(\Omega_C \approx 2\Delta\) defines the fraction of this weight which is transferred into condensate at \(T < T_c\). Therefore, the magnitude of \(\lambda_c\) can be expected to systematically decrease with the enhancement of the DC conductivity, in accord with the FGT sum rule. Notably, an approximate form (Eq. 2) yields the \(\lambda_c(\sigma_{DC})\) scaling with the power law \(\alpha = 1/2\) which is close to \(\alpha = 0.59\) seen in Fig. 1.

Interestingly, the sum rule arguments discussed above successfully describe the universal \(\lambda_c - \sigma_{DC}\) scaling in the underdoped cuprates despite the fact that the superconducting energy gap is not well-defined in the interlayer conductivity of these materials. The gap-less response of underdoped cuprates is exemplified in left panel in Fig. 2 displaying \(\sigma_1(\omega)\) data for Pr_{0.3}Y_{0.7}Ba_2Cu_{3}O_{6.95} with \(T_c \approx 60\) K. The conductivity was determined through a Kramers-Kronig analysis of reflectivity measured over the frequency range from 10 cm\(^{-1}\) to 48,000 cm\(^{-1}\). Regardless of the low- and high-energy extrapolations, the \(\sigma_1(\omega)\) data is unchanged over the energy interval displayed in Fig. 2. The response of the Pr-doped sample is similar to the features found in the conductivity of under-doped oxygen deficient YBa_2Cu_3O_7-\(\delta\) (YBCO) \(\sigma_1(\omega)\). Namely, the normal state conductivity is suppressed as the sample is cooled down to \(T_c\), with a transfer of spectral weight to higher energies. Below \(T_c\), one does not find any radical changes in the \(\sigma_1(\omega)\) spectra of Pr-doped YBCO or in the oxygen-deficient YBCO with a similar \(T_c\). Most importantly, this system, along with all other under-doped compounds, shows significant absorption in the superconducting state so that \(\sigma^{SC}_{DC} > 0\). Therefore, in cuprates only a small fraction of the far-infrared spectral weight is contributing to the condensate. The latter conclusion is in apparent conflict with the assumption \(\sigma^{SC}_{DC}(\omega < 2\Delta) \approx 0\), which allows one to reduce Eq. 3 to an approximate form given by Eq. 4. However, the strong condensate density seen in the cuprates located on the universal line can be understood in terms of the dramatic enhancement of the energy scale \(\Omega_C\) over the magnitude of the energy gap. This latter conclusion also follows from the explicit sum rule analysis for samples of underdoped La_{2-x}Sr_xCuO_4 (La214) and YBCO materials, suggesting that \(\Omega_C\) in these compounds exceeds 0.1-0.2 eV \(\sigma_1(\omega)\).

The involvement of a broad energy scale into the formation of the condensate in underdoped cuprates is also supported through a comparison of the universal scaling patterns seen for these materials and of a similar pattern detected for non-cuprate superconductors. According to Eqs. 3 and 4, the energy scale associated with the condensate formation for materials on the upper line is of the order of 3 meV which is close to estimates of the gap for most "conventional" materials in Fig. 1. If Eq. 2 is employed to describe the difference between the upper and the lower lines in Fig. 3, than one is forced to conclude that the corresponding scale for underdoped cuprates is of the order of \(\sim 0.3\) eV. As pointed out above, this assessment of \(\Omega_C\) is supported by the explicit sum rule analysis \(\sigma_1(\omega)\) and also makes \(\Omega_C\) the largest energy scale in the problem of cuprate superconductivity. Data points in Fig. 3 for overdoped materials support the notion that the \(\lambda_c - \sigma_{DC}\) plot provides means to learn about the energy scale associated with the condensate formation. Deflection of the over-doped cuprates from the universal line implies that \(\Omega_C\) is gradually suppressed with the increased carrier density. This trend is common for Tl_2Ba_2CuO_{6+\delta} (Tl2201), La214 and YBCO materials (see Fig. 3). Integration of the conductivity for all these materials shows that the FGT sum rule is exhausted at energies as low as 0.08 eV \(\sigma_1(\omega)\).

A quick inspection of the materials in Fig. 1 suggests that the \(\sim 3\) meV scale is observed in systems in which superconductivity emerges out of a FL state whereas the enhanced value of \(\Omega_C \approx 0.3\) eV is found in non-FL underdoped cuprates. The experiments which in our opinion are most relevant to FL versus non-FL classification include quantum oscillations of the low-\(T\) inter-layer resistivity (and of other quantities) in high magnetic fields \(\sigma_1(\omega)\). Quantum oscillations is a direct testimony of long-lived quasiparticles which are capable of propagating coherently between the layers. Such quasiparticles are a prerequisite of the Fermi-liquid ground state. On the contrary, quantum oscillations have never been reported for under-doped cuprates. The lack of coherence in the \(c\)-axis transport in these materials indicates that the ground state of cuprates is distinct from the FL picture.

Signatures of FL versus non-FL behavior also can be recognized in the spectra of the \(c\)-axis conductivity. A hallmark of the FL response is the Drude peak seen in \(\sigma_1(\omega)\) of metals. Notably, this feature is never found in underdoped compounds (forming the non-FL line in Fig. 3). The electronic contribution to \(\sigma_1(\omega)\) in these materials is usually structureless which is commonly associated with the incoherent (diffusive) motion of charge carriers across the planes. On the contrary, many materials that belong to the upper (FL) line in Fig. 3 demonstrate a familiar Drude behavior. This behavior has been found in Sr_2RuO_4 \(\sigma_1(\omega)\) and also shown in our data for inter-plane response of NbSe_2 (Fig. 2, right panel), reported here for the first time. In both cases, the width of the peak decreases at low temperatures, which is characteristic of the response of ordinary metals. Earlier measurements of the \(c\)-axis thermo reflectance proved applicability of conventional BCS electrodynamics to the NbSe_2 data \(\sigma_1(\omega)\). As for the over-doped compounds (located in a cross-over region between non-FL and FL lines in Fig. 3) their conductivity is indicative of the formation of the Drude peak (Fig. 2, middle panel), which is becoming more pronounced with the increase of the carrier density.
To summarize the experimental results, we wish to stress the following points: i) two distinct patterns in $\lambda - \sigma_{DC}$ correlation (Fig. 1) originate from a dramatic difference ($\simeq 10^2$) in the energy scale $\Omega_C$ from which the interlayer condensate is collected; ii) the pattern with the typical energy scale of $\simeq 3$ meV is realized in the materials with the coherent FL-type transport between the planes, whereas the one with $\Omega \simeq 300$ meV is found in cuprate superconductors with the incoherent non-FL response; iii) over-doped cuprates reveal a cross-over between the two behaviors. These results allow us to draw several conclusions regarding the features of the superconducting condensate in different layered systems.

- The symmetry of the order parameter seems to be unrelated to trends seen in the condensate response. Indeed, the upper line in Fig. 1 is formed by $s$-wave transition metal dichalcogenides, $p$-wave Sr$_2$RuO$_4$ and organic materials for which both $s$- and $d$-wave states have been proposed [11], while $d$-wave high-$T_c$ materials form the lower line and the crossover region between the lines.

- Electrodynamic of the FL systems at $T \ll T_c$ is determined by the magnitude of the gap (and hence by $T_c$), in general agreement with the BCS theory. It is therefore hardly surprising that the trend initiated by layered FL materials is also followed in 1-dimensional organic conductors as well as by more conventional systems such as Nb Josephson junctions, bulk Nb, Pb or amorphous aMo$_1$-$x$Ge$_x$ (see Fig. 1).

- We find no obvious connection between the broad energy scale $\Omega_C$ and the critical temperature of the studied superconductors. While scaling of $\lambda_c$ by the magnitude of $T_c$ does reduce the "scattering" of the data points, the two distinct $\lambda_c - \sigma_{DC}$ patterns persist even if such scaling is implemented. In particular, the critical temperature of under-doped La$_{214}$ materials is nearly the same as that of the several ET-compounds ($\simeq 12 - 15$ K). Nevertheless, the penetration depth is dramatically enhanced in the latter system.

- While the pseudogap state was shown to be responsible for the anomalous superfluid response of the underdoped cuprates [10], the characteristic pseudogap temperature $T^* = 90 - 350$ K is still much lower than our estimate of $\Omega_C$ for these materials. Dramatic enhancement of the $\Omega_C$ scale well beyond the gap value in underdoped cuprates may be connected with a well-established feature of the electronic spectral function $A(\omega)$ in underdoped cuprates. At $T > T_c$ the spectral function shows no quasiparticle peak in the antinodal direction; the spectral weight of $A(\omega)$ appears primarily in the incoherent channel and is spread out over the energy interval extending beyond 0.3-0.5 eV [13]. The coherent contribution to $A(\omega)$ is observed in the superconducting state and the $T$-dependence of its weight is similar to that of the superfluid density. [10] These experiments suggest that the emergence of the condensate is associated with the changes in the incoherent region of the spectral function. We stress a quantitative agreement between our estimate of the $\Omega_C$ and the energy region affected at $T < T_c$ in the ARPES measurements. [8]

Because carrier condensation is connected with the perturbation of the entire spectral function, the scale set by the superconducting gap becomes irrelevant to the problem of the interplane electrodynamics of underdoped cuprates in accord with our findings. The development of coherence in the spectral function at $T < T_c$ may be consistent with the lowering of the electronic kinetic energy [5], a result which was inferred earlier from the sum rule analysis of the interlayer conductivity [4].

Close correspondence between the properties of the spectral function in the $(\pi, 0)$ region and the features of the interlayer conductivity may stem from the peculiarities of the interlayer transfer matrix elements which are maximized for this particular region [14]. Therefore it is natural to look for a common microscopic origin of the anomalies in the $c$-axis response and characteristic features of the ARPES spectra at $(\pi, 0)$. The latter has been associated with the electron fractionalization at $T > T_c$ [20]. An important attribute of scenarios of cuprate superconductivity discussed in Ref. [20] is that at $T < T_c$ the total energy is lowered due to reduction of the kinetic energy, in qualitative agreement with the data. A quantitative account of the distinct energy scales associated with the condensate in non-FL materials is a challenge for models attempting to describe superconductivity in the cuprates. This research was supported by the U.S. DOE, NSF and Research Corporation.

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FIG. 1. The $c$-axis penetration depth reveals two patterns of scaling behavior between the magnitude of the interlayer penetration depth $\lambda_c(T=0K)$ and DC-conductivity $\sigma_{DC}(T)$. Most cuprate superconductors exhibit much shorter penetration depths than non-cuprates materials with the same $\sigma_{DC}(T_c)$. This result implies dramatic enhancement of the energy scale $\Omega_c$ from which the condensate is collected as described in the text. Data points: YBCO [4,21,22], overdoped YBCO [4,8,23], La214 [5,22,24], HgBa2Cu3O4 [25], Ti2201 [2,3,9], Bi2Sr2CaCu2O8 [26], Nd2−xCexCuO4 [27]. Blue points - underdoped, green - optimally doped; red - overdope. Transition metal dichalcogenides [13], (ET)2X compounds [28], (TMTSF)2ClO4 [29], Sr2RuO4 [30], niobium [31,32], lead [32], niobium Josephson junctions [33] and αMo1−xGe2 [34]. Inset: in a conventional dirty limit superconductor the spectral weight of the superconducting condensate (given by 1/$\lambda^2$) is collected primarily from the energy gap region. The total normal weight is preset by magnitude of $\sigma_{DC}$ whereas the product of $\Delta \times \sigma_{DC}$ quantifies the fraction of the weight that condenses.

FIG. 2. Examples of the interplane optical conductivity for layered superconductors. The observation of the Drude feature in the interplane optical conductivity of the dichalcogenide 2H-NbSe2 (right panel) is consistent with magnetoresistance measurements that revealed evidence for well-behaved quasiparticles. Instead, the conductivity of the underdoped Pr0.95Y0.05Ba2Cu3O8 material (left panel) suggests no signs of coherent response. Overdoped cuprates show the emergence of the Drude feature (middle panel) and also occupy intermediate position between non-FL and FL data set in Fig. 1.
Figure 1

![Diagram showing various compounds and their properties](attachment:image.png)

- α-(ET)$_2$NH$_4$Hg(SCN)$_4$
- β"-(ET)$_2$SF$_5$CH$_2$CF$_2$SO$_3$
- κ-(ET)$_2$I$_3$
- TaS$_2$(py)$_{1/2}$
- κ-(ET)$_2$Cu(NCS)$_2$
- κ-(ET)$_2$Cu[N(CS)$_2$]Br
- (TMTSF)$_2$ClO$_4$
- α-(ET)$_2$Mo$_{1-x}$Ge$_x$
- NbSe$_2$
- Sr$_2$RuO$_4$
- α-Mo$_{1-x}$Ge$_x$
- Nb/Nb
- Nb/Pb

The figure compares the λ$_c$ and σ$_1$ values for various superconducting materials, including YBCO, LSCO, Tl 2201, Hg 1201, Bi 2212, NCCO, Pr-YBCO, and others. The y-axis represents λ$_c$ in [μm], and the x-axis represents σ$_1$ in [Ω$^{-1}$ cm$^{-1}$]. The inset shows σ$_1$(ω) at T = T$_c$ and T = 0 K. The shaded area indicates the range of σ$_1$ for different materials.
Figure 2