Two gaps make a high-temperature superconductor?

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Abstract
One of the keys to the high-temperature superconductivity puzzle is the identification of the energy scales associated with the emergence of a coherent condensate of superconducting electron pairs. These might provide a measure of the pairing strength and of the coherence of the superfluid, and ultimately reveal the nature of the elusive pairing mechanism in the superconducting cuprates. To this end, a great deal of effort has been devoted to investigating the connection between the superconducting transition temperature $T_c$ and the normal-state pseudogap crossover temperature $T^*$. Here we present a review of a large body of experimental data which suggests a coexisting two-gap scenario, i.e. superconducting gap and pseudogap, over the whole superconducting dome. We focus on spectroscopic data from cuprate systems characterized by $T_{\text{max}}^c \sim 95$ K, such as Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$, YBa$_2$Cu$_3$O$_{7-\delta}$, Tl$_2$Ba$_2$CuO$_{6+\delta}$ and HgBa$_2$CuO$_{4+\delta}$, with particular emphasis on the Bi-compound which has been the most extensively studied with single-particle spectroscopies.

Some figures in this article are in colour only in the electronic version.

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1. Introduction
Since their discovery \cite{1}, the copper-oxide high-$T_c$ superconductors (HTSCs) have become one of the most investigated class of solids \cite{2–24}. However, despite the intense theoretical and experimental scrutiny, an understanding of the mechanism that leads to superconductivity is still lacking. At the very basic level, what distinguishes the cuprates from the conventional superconductors is the fact that they are doped materials, the highly atomic-like Cu 3d orbitals give rise to strong electron correlations (e.g. the undoped parent compounds are antiferromagnetic Mott–Hubbard-like insulators), and the superconducting elements are weakly-coupled two-dimensional layers (i.e. the celebrated square CuO$_2$ planes). Among the properties that are unique to this class of superconducting materials, in addition to the unprecedented high superconducting $T_c$, the normal-state gap or pseudogap is perhaps the most noteworthy. The pseudogap was first detected in the temperature dependence of the spin-lattice relaxation and Knight shift in nuclear magnetic resonance and magnetic susceptibility studies \cite{25}. The Knight shift is proportional to the density of states at the Fermi energy; a gradual depletion was observed below a crossover temperature $T^*$, revealing the opening of the pseudogap well above $T_c$ on the underdoped side of the HTSC phase diagram (figure 1). As the estimates based on thermodynamic

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quantities are less direct than in spectroscopy, we, in the course of this review, concentrate mainly on spectroscopic results; more information on other techniques can be found in the literature [5].

As established by a number of spectroscopic probes, primarily angle-resolved photoemission spectroscopy, [26,27] the pseudogap manifests itself as a suppression of the normal-state electronic density of states at $E_F$ exhibiting a momentum dependence reminiscent of a $d_{x^2−y^2}$ functional form. For hole-doped cuprates, it is largest at Fermi momenta close to the antinodal region in the Brillouin zone— i.e. around $(\pi,0)$— and vanishes along the nodal direction— i.e. the $(0,0)$ to the $(\pi,\pi)$ line. Note however that, strictly speaking, photoemission and tunneling probe a suppression of spectral weight in the single-particle spectral function, rather than directly of density of states; to address this distinction, which is fundamental in many-body systems and will not be further discussed here, it would be very interesting to investigate the quantitative correspondence between nuclear magnetic resonance and single-particle spectroscopy results. Also, no phase information is available for the pseudogap since, unlike the case of optimally and overdoped HTSCs [28], no phase-sensitive experiments have been reported for the underdoped regime where $T^* \gg T_c$.

As for the doping dependence, the pseudogap $T^*$ is much larger than the superconducting $T_c$ in underdoped samples, it smoothly decreases upon increasing the doping, and seems to merge with $T_c$ in the overdoped regime, eventually disappearing together with superconductivity at doping levels larger than $x \sim 0.27$ [5–24].

In order to elaborate on the connection between pseudogap and high-$T_c$ superconductivity, or in other words between the two energy scales $E_{pg}$ and $E_{sc}$ identified by $T^*$ and $T_c$, respectively, let us start by recalling that in conventional superconductors the onset of superconductivity is accompanied by the opening of a gap at the chemical potential in the one-electron density of states. According to the Bardeen–Cooper–Schrieffer (BCS) theory of superconductivity [29], the gap energy provides a direct measure of the binding energy of the two electrons forming a Cooper pair (the two-particle bosonic entity that characterizes the superconducting state). It therefore came as a great surprise that a gap, i.e. the pseudogap, was observed in the HTSCs not only in the superconducting state as expected from BCS, but also well above $T_c$. Because of these properties and the hope it might reveal the mechanism for high-temperature superconductivity, the pseudogap phenomenon has been very intensely investigated. However, no general consensus has been reached yet on its origin, its role in the onset of superconductivity itself, and not even on its evolution across the HTSC phase diagram.

As discussed in three recent papers on the subject [12,15,17], and here summarized in figure 1, three different phase diagrams are usually considered with respect to the pseudogap line. While Millis [15] opts for a diagram like the one in figure 1(a), Cho [17] prefers a situation where the pseudogap line meets the superconducting dome at $x \simeq 0.16$ (figures 1(b) and (c)); Norman et al [12] provide a comprehensive discussion of the three different possibilities. One can summarize some of the key questions surrounding the pseudogap phenomenon and its relevance to high-temperature superconductivity as follows [12, 15, 17]:

1. Which is the correct phase diagram with respect to the pseudogap line?
2. Does the pseudogap connect to the insulator quasiparticle spectrum?
3. Is the pseudogap the result of some one-particle band structure effect?
4. Or, alternatively, is it a signature of a two-particle pairing interaction?
5. Is there a true order parameter defining the existence of a pseudogap phase?
6. Do the pseudogap and a separate superconducting gap coexist below $T_c$?
7. Is the pseudogap a necessary ingredient for high-$T_c$ superconductivity?

In this review we revisit some of these questions, with specific emphasis on the one- versus two-gap debate. Recently, this latter aspect of the HTSCs has been discussed in great detail by Goss Levi [30], in particular based on scanning-tunneling microscopy data from various groups [31–33]. Here we expand this discussion to include the plethora of experimental results available from a wide variety of techniques. We

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**Figure 1.** Various scenarios for the interplay of pseudogap (blue dashed line) and superconductivity (red solid line) in the temperature-doping phase diagram of the HTSCs. While in (a) the pseudogap merges gradually with the superconducting gap in the strongly overdoped region, in (b) and (c) the pseudogap lines intersect the superconducting dome at about optimal doping (i.e. maximum $T_c$). In most descriptions, the pseudogap line is identified with a crossover with a characteristic temperature $T^*$ rather than a phase transition; while at all dopings $T^* > T_c$ in (a), beyond optimal doping $T^* < T_c$ in (b) and $T^*$ does not even exist in (c). Adapted from [12].
show that one fundamental and robust conclusion can be drawn: the HTSC phase diagram is dominated by two energy scales, the superconducting transition temperature $T_c$ and the pseudogap crossover temperature $T^*$, which converge to the same critical point at the end of the superconducting dome. Establishing whether this phenomenology can be conclusively described in terms of a coexisting two-gap scenario, and what the precise nature of the gaps would be, will require a more definitive understanding of the quantities measured by the various probes.

2. Emerging phenomenology

The literature on the HTSC superconducting gap and/or pseudogap is very extensive and still growing. In this situation it seems interesting to go over the largest number of data obtained from as many experimental techniques as possible, whenever possible. With respect to the spectroscopic data, it is important to differentiate between single-particle probes such as ARPES and STM, which directly measure the one-electron excitation energy $\Delta$ with respect to the chemical potential (on both side of $E_F$ in STM), and two-particle probes such as Raman and inelastic neutron scattering, which instead provide information on the particle-hole excitation energy $2\Delta$. Note that the values reported here are those for the ‘full gap’ $2\Delta$ (associated with either $E_{sc}$ or $E_{pg}$), while frequently only half the gap $\Delta$ is given for instance in the ARPES literature. In doing so one implicitly assumes that the chemical potential lies half-way between the lowest-energy single-electron removal and addition states; this might not necessarily be correct but appears to be supported by the direct comparison between ARPES and STM/Raman results. A more detailed discussion of the quantities measured by the different experiments and their interpretation is provided in the following subsections. Here we would like to point out that studies of $B_{2g}$ and $B_{1g}$ Raman intensity [19, 40, 52], heat conductivity of nodal quasiparticles [70, 71] and neutron magnetic resonance energy $\Omega_2$ [42] do show remarkable agreement with superconducting or pseudogap energy scales as inferred by single-particle

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**Figure 2.** Pseudogap ($E_{pg} = 2\Delta_{pg}$) and superconducting ($E_{sc} \sim 5k_BT_c$) energy scales for a number of HTSCs with $T_{c\text{max}} \sim 95$ K (Bi2212, Y123, Tl2201 and Hg1201). The datapoints were obtained, as a function of hole doping $x$, by angle-resolved photoemission spectroscopy (ARPES), tunneling (STM, SIN, SIS), Andreev reflection (AR), Raman scattering (RS) and heat conductivity (HC). On the same plot we are also including the energy $\Omega_2$, of the magnetic resonance mode measured by inelastic neutron scattering (INS), which we identify with $E_{pg}$ because of the striking quantitative correspondence as a function of $T_c$. The data fall on two universal curves given by $E_{pg} = E_{pg}^\text{max}(0.27 - x)/0.22$ and $E_{sc} = E_{sc}^\text{max}[1 - 82.6(0.16 - x)^2]$, with $E_{pg}^\text{max} = E_{pg}(x = 0.05) = 152 \pm 8$ meV and $E_{sc}^\text{max} = E_{sc}(x = 0.16) = 42 \pm 2$ meV (the statistical errors refer to the fit of the selected datapoints; however, the spread of all available data would be more appropriately described by ±20 and ±10 meV, respectively).
probes, or with the doping dependence of $T_c$ itself. Thus they provide, in our opinion, an additional estimate of $E_{sc}$ and $E_{pg}$ energy scales.

As for the choice of the specific compounds to include in our analysis, we decided to focus on those HTSCs exhibiting a similar value of the maximum superconducting transition temperature $T_{c,max}$, as achieved at optimal doping, so that the data could be quantitatively compared without any rescaling. We have therefore selected Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212), YBa$_2$Cu$_3$O$_{6+\delta}$ (Y123), Tl$_2$Ba$_2$CuO$_{6+\delta}$ (Tl2201) and HgBa$_2$CuO$_{4+\delta}$ (Hg1201), which have been extensively investigated and are all characterized by $T_{c,max} \sim 95$ K [73] (with particular emphasis on Bi2212, for which the most extensive set of single-particle spectroscopy data is available). It should also be noted that while Bi2212 and Y123 are ‘bilayer’ systems, i.e. their crystal structure contains as a key structural element sets of two adjacent CuO$_2$ layers, Tl2201 and Hg1201 are structurally simpler single CuO$_2$-layer materials. Therefore, this choice of compounds ensures that our conclusions are generic to all HTSCs with a similar $T_c$, independent of the number of CuO$_2$ layers.

A compilation of experimental results for the magnitude of pseudogap ($E_{pg} = 2\Delta_{pg}$) and superconducting ($E_{sc} \sim 5k_B T_c$) energy scales, as a function of carrier doping $x$, is presented in figure 2 (only some representative datapoints are shown, as not to overload the figure; similar compilations were also obtained by a number of other authors) [5, 9, 13, 16, 42, 43, 52, 57, 60, 70, 74, 75]. The data for these HTSCs with comparable $T_{c,max} \sim 95$ K fall on two universal curves: a straight line for the pseudogap energy $E_{pg} = 2\Delta_{pg}$ and a parabola for the superconducting energy scale $E_{sc} \sim 5k_B T_c$. The two curves converge to the same $x \sim 0.27$ critical point at the end of the superconducting dome, similarly to the cartoon of figure 1(a). In order to summarize the situation with respect to quantitative estimates of $E_{pg}$ and $E_{sc}$, we have listed in table 1 the values as determined by the different experimental techniques on optimally doped Bi2212 (with $T_c$ ranging from 90 to 95 K). While one obtains from this compilation the average values of $E_{pg} \simeq 76$ meV and $E_{sc} \simeq 41$ meV at optimal doping, the numbers do scatter considerably. Note also that these numbers differ slightly from those given in relation to the parabolic and straight lines in figure 2 (e.g. $E_{sc}^{max} = 42$ meV) because the latter were inferred from a fitting of superconducting and pseudogap data over the whole doping range, while those in table 1 were deduced from results for optimally doped Bi2212 only. It is also possible to plot the pseudogap $E_{pg}$ and superconducting $E_{sc}$ energy scales as estimated simultaneously in one single experiment on the very same sample. This is done in figure 3 for Raman, tunneling and ARPES results from Bi2212 and Hg1201, which provide evidence for the presence of two energy scales, or possibly two spectral gaps as we discuss in greater detail below, coexisting over the whole superconducting dome.

### 2.1. Angle-resolved photoemission

The most extensive investigation of excitation gaps in HTSCs has arguably been done by ARPES [9, 10, 26, 27, 34, 35, 54–66, 76–80]. This technique provides direct access to the one-electron removal spectrum of the many-body system; it allows, for instance in the case of a BCS superconductor [29], to measure the momentum dependence of the absolute value of the pairing amplitude $2\Delta$ via the excitation gap $\Delta$ observed for single-electron removal energies, again assuming $E_F$ to be located half-way in the gap [9, 10]. This is the same in some tunneling experiments such as STM, which however do not provide direct momentum resolution but measure on both sides of $E_F$ [18]. The gap magnitude is usually inferred from the ARPES spectra from along the normal-state Fermi surface in the antinodal region, where the $d$-wave gap is largest; it is estimated from the shift to high-binding energy of the quasiparticle spectral weight relative to the Fermi energy. With this approach only one gap is observed below a temperature scale that smoothly evolves from the so-called pseudogap temperature $T^* \sim 50$ K in the underdoped regime, to the superconducting $T_c$ on the overdoped side. We identify this gap as $E_{pg}$ and not $E_{sc}$. The two energy scales, or possibly two spectral gaps, are evidently coexisting over the whole superconducting dome.

### Table 1.

| Experiment | Energy | meV | References |
|------------|--------|-----|------------|
| ARPES—$(\pi,0)$ peak | $E_{pg}$ | 80 | [34, 35] |
| Tunneling—STM | | 70 | [18, 36] |
| Tunneling—SIN | | 85 | [37] |
| Tunneling—SIS | | 75 | [38, 39] |
| Raman—$B_{1g}$ | | 65 | [40] |
| Electrodymanics | | 80 | [5, 41] |
| Neutron—$(\pi, \pi)$ | $E_{sc}$ | 40 | [42] |
| Raman—$B_{2g}$ | | 45 | [40] |
| Andreev | | 45 | [43] |
| SIS—dip | | 40 | [39] |

Figure 3. Pseudogap $E_{pg}$ and superconducting $E_{sc}$ energy scales ($2\Delta$) as estimated, by a number of probes and for different compounds, in one single experiment on the very same sample. These data provide direct evidence for the simultaneous presence of two energy scales, possibly two spectral gaps, coexisting in the superconducting state. The superconducting and pseudogap lines are defined as in figure 2.
with the pseudogap energy scale $E_{pg} = 2\Delta_{pg}$. This is also in agreement with recent investigations of the near-nodal ARPES spectra from single and double layer Bi-cuprates [57, 76, 77], which further previous studies of the underdoped cuprates’ Fermi arc phenomenology [78–80]. From the detailed momentum dependence of the excitation gap along the Fermi surface contour, and the different temperature trends observed in the nodal and antinodal regions, these studies suggest the coexistence of two distinct spectral gap components over the whole superconducting dome: superconducting gap and pseudogap, dominating the response in the nodal and antinodal regions, respectively, which would eventually collapse to one single energy scale in the very overdoped regime.

2.2. Tunneling

The HTSCs have been investigated by a wide variety of tunneling techniques [13, 18, 36–39, 44–51], such as SIN [38, 51], SIS [37–39], STM [18, 36, 46], intrinsic tunneling [47–50] and Andreev reflection, which is also a tunneling experiment but involves two-particle rather than single-particle tunneling (in principle, very much like SIS) [13, 43, 72]. All these techniques, with the exception of intrinsic tunneling, are represented here either in the figures or table.

Similarly to what was discussed for ARPES at the antinodes, there are many STM studies that report a pseudogap $E_{pg}$ smoothly evolving into $E_{sc}$ upon overdoping [18, 31]. In addition, a very recent temperature-dependent study of overdoped single-layer Bi-cuprate detected two coexisting, yet clearly distinct, energy scales in a single STM experiment [32]. In particular, while the pseudogap was clearly discernible in the differential conductance exhibiting the usual large spatial modulation, the evidence for a spatially uniform superconducting gap was obtained by normalizing the low-temperature spectra by those just above $T_c \approx 15$ K. These values have not been included in figures 2 and 3 because $T_c \ll 95$ K; however, this study arguably provides the most direct evidence for the coexistence of two distinct excitation gaps in the HTSCs.

One can regard Andreev reflection (pair creation in addition to a hole) as the inverse of a two-particle scattering event (with the magnetic scattering from a spin 1/2 system) [8, 16, 24, 42, 68, 69]; this weakens the magnetic response should be part of the considerations in the modeling of magnetic resonance mediated high-$T_c$ superconductivity. Alternatively, its detection below $T_c$ might be a mere consequence of the onset of superconductivity and of the corresponding suppression of quasiparticle scattering. Independently of the precise interpretation, the INS data reproduced in figure 2 show that the magnetic resonance energy $\Omega$ tracks very closely, over the whole superconducting dome, the superconducting...
energy scale $E_{\text{sc}} \sim 5k_{\text{B}}T_{\text{c}}$ (similar behavior is observed, in the underdoped regime, also for the spin-gap at the incommensurate momentum transfer $(\pi, \pi \pm \delta)$ [81]). Also remarkable is the correspondence between the energy of the magnetic resonance and that of the $B_{2g}$ Raman peak. Note that while the $q = (\pi, \pi)$ momentum transfer observed for the magnetic resonance in INS is a key ingredient of most proposed HTSC descriptions, Raman scattering is a $q = 0$ probe. It seems that understanding the connection between Raman and INS might reveal very important clues.

2.5. Heat conductivity

Heat conductivity data from Y123 and Tl2201 fall onto the pseudogap line. This is a somewhat puzzling result because they have been measured at very low temperatures, well into the superconducting state, and should in principle provide a measure of both gaps together if these were indeed coexisting below $T_{\text{c}}$. However, similarly to the $B_{2g}$ Raman scattering, these experiments are only sensitive to the slope of the gap function along the Fermi surface at the nodes, $(1/\hbar)(d\Delta/dq)_{\text{sc}}$; the gap itself is determined through an extrapolation procedure in which only one gap was assumed. The fact that the gap values, especially for Y123, come out on the high side of the pseudogap line may be an indication that an analysis with two coexisting gaps might be more appropriate.

3. Outlook and conclusion

The data in figures 2 and 3 demonstrate that there are two coexisting energy scales in the HTSCs: one associated with the superconducting $T_{\text{c}}$ and the other, as inferred primarily from the antinodal region properties, with the pseudogap $T^*$. The next most critical step is that of addressing the subtle questions concerning the nature of these energy scales and the significance of the emerging two-gap phenomenology towards the development of a microscopic description of high-$T_{\text{c}}$ superconductivity.

As for the pseudogap, which grows upon underdoping, it seems natural to seek a connection to the physics of the insulating parent compound. Indeed, it has been pointed out that this higher energy scale might smoothly evolve, upon underdoping, into the quasiparticle dispersion observed by ARPES in the undoped antiferromagnetic insulator [82, 83]. At zero doping the dispersion and quasiparticle weight in the single-hole spectral function as seen by ARPES can be very well explained in terms of a self-consistent Born approximation [84], as well as in the diagrammatic quantum Monte Carlo [85] solution to the so-called $t$-$t'$-$t''$-$J$ model. In this model, as in the experiment [82, 83], the energy difference between the top of the valence band at $(\pi/2, \pi/2)$ and the antinodal region at $(\pi, 0)$ is a gap due to the quasiparticle dispersion of about $250 \pm 30$ meV. Note that this would be a single-particle gap $\Delta$. For the direct comparison with the pseudogap data in figure 2, we would have to consider $2\Delta \sim 500$ meV; this, however, is much larger than the $x = 0$ extrapolated pseudogap value of 186 meV found from our analysis across the phase diagram. Thus there seems to be an important disconnection between the finite doping pseudogap and the zero-doping quasiparticle dispersion.

The fact that the pseudogap measured in ARPES and SIN experiments is only half the size of the gap in SIS, STM, $B_{1g}$ Raman and heat conductivity measurements, points to a pairing gap. So although the origin of the pseudogap at finite doping remains uncertain, we are of the opinion that it most likely reflects a pairing energy of some sort. To this end, the trend in figure 2 brings additional support to the picture discussed by many authors that the reduction in the density of states at $T^*$ is associated with the formation of electron pairs, well above the onset of phase coherence taking place at $T_{\text{c}}$ (see, e.g. [86, 87]). The pseudogap energy $E_{\text{pg}} = 2\Delta_{\text{pg}}$ would then be the energy needed to break up a preformed pair. To conclusively address this point, it would be important to study very carefully the temperature dependence of the $(\pi, 0)$ response below $T_{\text{c}}$; any further change with the onset of superconductivity, i.e. an increase in $E_{\text{pg}}$, would confirm the two-particle pairing picture, while a lack thereof would suggest a one-particle band structure effect as a more likely interpretation of the pseudogap.

The lower energy scale connected to the superconducting $T_{\text{c}}$ (parabolic curve in figure 2 and 3) has already been proposed by many authors to be associated with the condensation energy [86–89], as well as with the magnetic resonance in INS [90]. One might think of it as the energy needed to take a pair of electrons out of the condensate; however, for a condensate of charged bosons, a description in terms of a collective excitation, such as a plasmon or roton, would be more appropriate [24]. The collective excitation energy would then be related to the superfluid density and in turn to $T_{\text{c}}$. In this sense, this excitation would truly be a two-particle process and should not be measurable by single-particle spectroscopies. Also, if the present interpretation is correct, this excitation would probe predominantly the charge-response of the system; however, there must be a coupling to the spin channel, so as to make this process neutron active (yet not as intense as predicted by the sum rule for pure spin-$1/2$ magnetic excitations, which is consistent with the small spectral weight observed by INS). As discussed, one aspect that needs to be addressed to validate these conjectures is the surprising correspondence between $q = 0$ and $q = (\pi, \pi)$ excitations, as probed by Raman and INS, respectively.

We are led to the conclusion that the coexistence of two energy scales is essential for high-$T_{\text{c}}$ superconductivity, with the pseudogap reflecting the pairing strength and the other, always smaller than the pseudogap, the superconducting condensation energy. This supports the proposals that the HTSCs cannot be considered as classical BCS superconductors, but rather are smoothly evolving from the BEC into the BCS regime [91–93], as carrier doping is increased from the underdoped to the overdoped side of the phase diagram.

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