Interplay between the pseudogap and superconductivity in underdoped HgBa2CuO4+δ single crystals

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We report a doping dependant Electronic Raman Scattering (ERS) study of HgBa2CuO4+δ (Hg-1201) single crystals. We investigate the dynamics of the antinodal and nodal quasiparticles. We show that the dynamical response of the antinodal quasiparticles is strongly reduced towards the underdoped regime in both the normal and superconducting states. When probing the nodal quasiparticles, we are able to distinguish between the energy scale of the pseudogap and that of the superconducting gap. A simple model relating the suppression of the dynamical response of the antinodal quasiparticles to fluctuations related to a competing phase is proposed.

Among the unsolved problems of the high temperature superconductivity field, the question of the nature of the pseudogap phase is certainly the most highly debated. In particular its relationship with superconductivity remains an open question. One class of model attributes the pseudogap phase as a precursor phase of superconductivity in which Cooper pairs form at a temperature $T^*$ but only acquire phase coherence at a lower temperature $T_c$ where they form an uniform $d$-wave BCS condensate. An important consequence of all these theories is that the pseudogap and the superconducting gap are intimately connected: they can be both identified to a pairing energy. This approach is supported by ARPES data which show that the pseudogap and the superconducting gap open in the same region of the Fermi surface, i.e. close to the $(\pi,0)$ and related points. Another class of models invokes various phases which are not directly related to superconductivity but rather compete with it. Among the proposed phases we find a precursor SDW phase, a $d$-density wave phase or an orbital current phase. Most of these theories, but not all, predict the presence of a quantum critical point somewhere near the optimally doped regime. Specific heat data, doping evolution of the superfluid density and impurity induced $T_c$ suppression, among others, have been interpreted as evidences for this scenario.

In order to fully understand the nature of the pseudogap, a two-particle response function able to probe quasiparticle dynamics on different regions of the Brillouin zone would be useful. In this report we show that Electronic Raman Scattering (ERS) is such a probe and report a doping dependant study of the interplay between the pseudogap and superconductivity in HgBa2CuO4+δ (Hg-1201) single crystals. Being a two-particle probe, ERS is able to access the charge dynamics in both the normal and the superconducting states. Moreover, through the use of particular sets of incident and scattered polarizations, it is able to probe different regions of the Fermi surface, i.e. the nodal (along the $(0,0)$-$(\pi, \pi)$ direction) and antinodal (along the $(0,0)$-$(\pi, 0)$ direction) quasiparticles. With this unique ability, ERS is thus expected to give important information on the pseudogap nature which are not accessible via one-particle probes such as ARPES.

With only one CuO2 plane per unit cell and a pure tetragonal symmetry, Hg-1201 is a very attractive compound to study the intrinsic physics of the CuO2 plane in the underdoped regime. In particular underdoped Hg-1201 is closer to its fully stoichiometric phase ($\delta=0$) than optimally and over-doped Hg-1201 and should therefore be structurally more ordered. In this study, we show that the pseudogap in Hg-1201 manifests itself as a suppression of the superconducting Raman response along the antinodal directions. Comparison with available data on other compounds shows that this suppression is generic to the cuprates and starts slightly above optimal doping. When probing the nodal directions, we observe a partial loss of spectral weight in the response which starts around 700 cm$^{-1}$ in the normal state, while below $T_c$, the superconducting gap opens at about 200 cm$^{-1}$ thus indicating a non-superconducting origin of the pseudogap. We propose a simple model which relates the suppression of the antinodal dynamical response to two-particle vertex suppression arising from fluctuations of a competing phase.

The Hg-1201 single crystals studied have been successfully grown by the flux method. The detailed procedure for crystal growth will be described elsewhere. The ERS measurements have been performed on three as-grown single crystals with different dopings. The single crystals studied here were carefully selected from several batches for their sharp transitions widths (less than 5 K). Their magnetically measured transition temperatures, $T_c$, are 95 K, 78 K and 63 K. We will refer to these crystals as Hg95K, Hg78K and Hg63K respectively. The first sample is very close to optimal doping while the latter two are underdoped. The as-grown surfaces of the single crystals were mechanically polished to suppress small extrinsic impurity phases which are known to exist at the surface of mercurate compounds. The spectra were obtained
using the 514.5 nm (2.4 eV) excitation line of an Ar+-Kr+ laser. The scattered light was analysed using a triple grating spectrometer (JY-T64000) equipped with a nitrogen cooled CCD detector. In this study we focus on B1g and B2g symmetries which can be individually selected using specific configurations of incident and scattered electric fields with respect to the crystallographic axis. In the cuprates, the B1g (x’‘y’’ scattering geometry) symmetry is sensitive to regions along the antinodal directions while the B2g (xy scattering geometry) symmetry probes mostly along the nodal directions. The spectra presented here have been corrected for the spectral response of the spectrometer and for the Bose-Einstein factor. They are thus proportional to the imaginary part of the Raman response function $\chi''$. All the referred temperature have been corrected for the estimated laser heating.

In FIG. 1 we show the spectra in B1g and B2g symmetries as a function of doping both above and below $T_c$. The spectra for the optimally doped sample Hg95K are consistent with a d-wave superconducting gap and compare well with spectra of other optimally doped cuprates (for more details, see ref. 14). The B1g spectrum shows a redistribution into a 2$\Delta_0$ pair breaking peak upon entering the superconducting state (2$\Delta_0$ $\sim$ 520 cm$^{-1}$). On the other hand, the spectrum in the B2g symmetry displays a loss of spectral weight at low frequency but does not show any superconductivity induced peak. We note that for a d-wave gap, the B2g pair breaking peak intensity is expected to be much weaker than the B1g one and the peak should be located at a lower frequency, i.e. below 2$\Delta_0$. As we lower the doping, the response in the B1g symmetry displays a dramatic suppression of the pair breaking peak intensity and the normal and superconducting state spectra are virtually identical. By contrast, the B2g spectra show clear superconductivity induced peaks in both underdoped samples. The peaks are located around 360 cm$^{-1}$ and 200 cm$^{-1}$ for Hg78K and Hg63K respectively and are associated to the opening of the superconducting gap. It is well known that the B2g peak height is largely controlled by disorder.$^{10,15}$ The absence of a B2g peak in Hg95K is therefore most likely related to the fact that optimally doped Hg95K is structurally more disordered than the underdoped Hg78K and Hg63K. This is further confirmed by NMR measurements on Hg-1201 which show that $^{17}$O NMR linewidth increases when going from under to overdoped samples.$^{16}$

In fact it is remarkable that despite the complete disappearance of any coherent B1g superconducting response, the B2g response displays a clear coherent superconducting response for both underdoped samples.

In FIG. 2 the B1g and B2g responses of Hg63K are shown as a function of temperature. The first striking result is that the B1g spectra displays hardly any temperature dependence when cooling from 300 K down to 20 K. This suggests a highly incoherent response in this symmetry at least up to 1000 cm$^{-1}$. By contrast the B2g response displays a complex temperature dependence when cooling from 288 K to 20 K. Between 288 K and 215 K the response shows conventional metallic behavior, i.e. an overall increase of the spectral weight at low frequency. Between 215 K and 136 K, the response develops a partial suppression which starts below 700 cm$^{-1}$ and further deepens with cooling. The suppression is only partial because the 215 K and 136 K spectra cross again at about 130 cm$^{-1}$ consistently with a metallic-like behavior at very low frequency. We identify this suppression as the opening of an anisotropic pseudogap which leaves parts of the Fermi surface, along the nodal directions, essentially unaffected. A similar conclusion was reached by Nemetschek et al. from their analysis of the B2g response in underdoped Y-123 ortho II.$^{10}$ In the inset of FIG. 2 we plot the evolution of the integrated spectral weight (SW) as a function of temperature. The integrated SW increases until a characteristic temperature $T^*$ ($T^*$$\sim$170 K) where it starts to decrease until $T_c$ is reached. The deduced $T^*$ is very close to the one re-
ported by Itoh et al. based on their NMR data (631/T1T) on similarly doped Hg-1201 samples. The appearance of a pair breaking below Tc in the same symmetry allows for a direct comparison between the pseudogap and the superconducting gap. It shows very different associated energies: the pair breaking peak is located at 200 cm⁻¹ while the pseudogap opens around 700 cm⁻¹ thus advocating for a non-superconducting origin of the pseudogap.

We now discuss the striking evolution of the B1g pair breaking peak intensity with doping. The rapid disappearance of the B1g peak on the underdoped side of the phase diagram has also been reported for La-214, Bi-2212 and Y-123. Like the partial suppression observed in B2g symmetry, it was attributed to the presence of a pseudogap along the antinodal directions. More precisely, a destruction of the Fermi surface at the antinodal points was invoked. It is important however to stress that recent ARPES data show clear quasiparticle peaks at these points in the superconducting state of underdoped Bi(Pb)-2212, in apparent contradiction with Raman data in the superconducting state of underdoped cuprates. To quantify the suppression of the pair breaking peak intensity as we lower the doping, we have calculated the Superconducting Peak Ratio (SPR) defined as

\[
\text{SPR} = \frac{\chi''_S(\omega = 2\Delta_0)}{\chi''_N(\omega = 2\Delta_0)}
\]

where \(\chi''_S\) and \(\chi''_N\) are the Raman responses in the superconducting and normal states respectively. In FIG. 3 this ratio is plotted as a function of doping for various cuprates. The suppression is clearly a generic property of the hole-doped cuprates. In fact, the evolution of the SPR as a function of doping is strikingly similar to the evolution of the condensation energy and the c-axis superfluid density in various cuprates. FIG. 3 suggests that the pseudogap results from interactions which progressively destroy the superconducting response near the antinodal directions and leave the nodal directions unaffected at low energy. A similar picture has emerged recently from STM experiments where coexistence between nodal superconductivity and charge ordering involving antinodal quasiparticles was found.

In the following we consider a simple phenomenological model which can account for the absence of any coherent superconducting response in the B1g channel of underdoped cuprates. The Raman response is a two particle probe:

\[
\chi''(\omega) = \frac{2}{N} \sum_k \sum_{\Omega} \int d\omega \left[ f(\omega) - f(\omega + \Omega) \right] \frac{1}{\pi} \left[ (\hat{\Gamma}_k(\omega + \Omega), \hat{\Gamma}_k(\omega, \omega + \Omega)) \hat{\chi'}(\omega) \right]
\]

where \(\hat{\chi'}(\omega)\) is the full imaginary part of the Green’s function in the superconducting state. \(\hat{\Gamma}_k\) are the bare, renormalized Raman vertices, respectively, and \(\hat{\tau}_i\) are the Pauli matrices. If vertex corrections are ignored (\(\hat{\Gamma}_k = \gamma_k \hat{\tau}_i\)) the Raman response can be calculated from the knowledge of single particle properties such as the self-energy \(\Sigma_k\) and the superconducting gap \(\Delta_k\). While expression (1) has been used extensively to fit Raman spectra in optimally doped cuprates, the situation in the underdoped cuprates does not fit this simple picture since no pair breaking peak is observed in the B1g symmetry and the single particle properties deduced from ARPES are not strongly doping dependent. It appears therefore that the rapid decrease of the superconducting response in the B1g symmetry in the underdoped regime cannot be accounted by single particle properties alone.
Thus, in the following, we consider the effect of two-particle vertex renormalizations to $\hat{\Gamma}_k$. The renormalized vertex $\hat{\Gamma}_k$ may be calculated in a number of contexts corresponding to various competing phases. Due to the topology of the Fermi surface in hole doped cuprates, it can be shown that, while the $B_{2g}$ vertex amplitude is only mildly affected by charge suppressing fluctuations centered around $(\pi, \pi)$, the $B_{1g}$ vertex is strongly suppressed. These fluctuations may be related to antiferromagnetic (AF) order but various competing phases like the $d$-density wave phase are expected to induce a similar suppression.

The effect of these fluctuations on the Raman vertex can be modelled by taking the following phenomenological form for the renormalized Raman vertex: $\hat{\Gamma}_k = \gamma_k \Delta \exp(-\beta (\cos k_x a - \cos k_y a)^2)$ where $\beta$ is a dimensionless parameter which, in this framework, depends on the strength of the coupling between the anti-nodal quasiparticles and the charge suppressing fluctuations. The calculated responses for both channels, with and without vertex corrections, in the normal and superconducting states, are shown in FIG. 4. Contrary to the $B_{2g}$ response, the renormalized $B_{1g}$ response is strongly suppressed and the $2\Delta_0$ pair breaking peak becomes hardly detectable. The theoretical spectra are qualitatively consistent with the experimental data in underdoped cuprates and underline the crucial role of vertex corrections on the channel dependant Raman response.

In summary, we have shown that anisotropic two-particle vertex renormalizations probed by ERs demonstrate a dichotomy between the dynamics of nodal and anti-nodal electrons. Our ERs results augment prior ARPES work to show that manifestations of the pseudogap hints towards its non-superconducting origin.

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