The Pseudogap in La_{2-x}Sr_xCuO_4: A Raman Viewpoint

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We report the results of Raman scattering experiments on single crystals of La_{2-x}Sr_xCuO_4 (La214) as a function of temperature and doping. In underdoped compounds low-energy B_{2g} spectral weight is depleted in association with the opening of a pseudogap on regions of the Fermi surface located near $(\pm \pi, 0)$ and $(0, \pm \pi)$. The magnitude of the depletion increases with decreasing doping, and in the most underdoped samples, with decreasing temperature. The spectral weight that is lost at low-energies ($\omega \leq 800 \text{ cm}^{-1}$) is transferred to the higher energy region normally occupied by multi-magnon scattering. From the normal state B_{2g} spectra we have determined the scattering rate $\Gamma(\omega, T)$ of quasiparticles located near the diagonal directions in k-space. In underdoped compounds, $\Gamma(\omega, T)$ is suppressed at low temperatures for energies less than $E_g(x) \approx 800 \text{ cm}^{-1}$. The doping dependence of both the two-magnon scattering and the scattering rate suppression suggest that the pseudogap is characterized by an energy scale $E_g \sim J$, where $J$ is the antiferromagnetic superexchange energy. Comparison with the results from other techniques provides a consistent picture of the pseudogap in La214.

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I. INTRODUCTION

It is now clear [1] that the unusual electronic properties of the underdoped high temperature superconductors are strongly influenced by the presence of a normal state pseudogap (PG). However, the origin of this PG and any relation it might have to the occurrence of superconductivity, remain controversial issues. In an attempt to resolve these questions many recent experiments have attempted to determine the energy scale $E_g$, doping dependence, and symmetry associated with the PG. The experimental situation remains unclear, however, in that Raman [3,4,5], far infrared reflectivity (FIR) [6,7], specific heat [11] and some recent [12] angle resolved photoemission spectroscopy (ARPES) experiments yield $E_g \sim J$, where $J$ is the antiferromagnetic (AFM) exchange energy, while tunneling [13] and earlier ARPES experiments [4,7] on Bi$_2$Sr$_2$CaCu$_2$O$_y$ (Bi2212) found $E_g \sim \Delta$, the superconducting gap energy. The results of ARPES experiments [4,7] also suggest that the PG has conventional d-wave symmetry while Raman results [3] indicate that the gap is more localized to regions of the Fermi surface (FS) located near the axes in reciprocal space. Finally FIR [6,7], ARPES [4,7,13], and NMR [14,15] experiments indicate that at certain doping levels the PG is characterized by an onset temperature which is not well defined in specific heat [11,17] and some Raman measurements [12].

In an attempt to reconcile some of the apparent conflicts mentioned above we have carried out a systematic Raman investigation of the dependence of the PG parameters on doping and temperature in La_{2-x}Sr_xCuO_4 (La214). This compound is an excellent material for these studies for several reasons. It has a relatively simple structure with a single CuO$_2$ plane in the primitive cell. Thus the results are not influenced by structural complications such as those introduced by the chains in YBa$_2$Cu$_3$O$_y$ (Y123), or the structural modulations in Bi2212. Furthermore, the hole concentration is determined simply by the Sr concentration if oxygen stoichiometry is maintained. As a result, one can obtain high quality, well characterized single crystals of La214 that enable one to study the systematic evolution of the electronic properties throughout the complete doping range. The samples studied in this work were grown by a traveling floating-zone method [18] and were carefully characterized using x-ray diffraction, transport and susceptibility measurements [18]. The physical parameters of the La214 samples are summarized in (Table I).

| La$_{2-x}$Sr$_x$CuO$_4$ | Sr content (x) | $T_c$ (K) |
|--------------------------|----------------|---------|
| Underdoped               | 0.08           | 16      |
| Underdoped               | 0.11           | 27      |
| Underdoped               | 0.13           | 35      |
| Optim-doped              | 0.17           | 37      |
| Overdoped                | 0.19           | 32      |
| Overdoped                | 0.22           | 30      |

TABLE I. The physical parameters characterizing the samples studied in this paper. $T_c$ was determined magnetically and x is the nominal Sr concentration (Ref. 18).
We have previously investigated the B<sub>1g</sub> and B<sub>2g</sub> Raman spectra of La214 for doping levels 0.13 ≤ x ≤ 0.22, and found that in underdoped materials there is a significant depletion of B<sub>1g</sub> spectral weight at low-energies (ω ≤ 800 cm<sup>-1</sup>). Furthermore, the strength of this depletion increases rapidly as the doping level is decreased below optimum. In the La214 samples studied previously, and in other cuprates, the strength of the depletion was found to be approximately independent of temperature for T ≤ 300K. Finally, in optimally doped compounds the B<sub>1g</sub> spectra undergo a strong renormalization that results in the formation of a 2∆ peak for T ≤ T<sub>c</sub>. In all the underdoped cuprates, however, the B<sub>1g</sub> spectrum is unaffected by the superconducting transition at T<sub>c</sub>. In contrast to the significant doping induced changes that occur in the B<sub>1g</sub> spectra, the strength of the B<sub>2g</sub> spectra appear to be approximately independent of doping. In addition, a superconductivity induced renormalization is observed in the B<sub>2g</sub> channel for all doping levels.

In this paper we have extended our range of investigation of La214 to lower doping levels (x = 0.08 and x = 0.11) and to higher energies (ω ≤ 4000 cm<sup>-1</sup>). Spectra have been obtained in both B<sub>1g</sub> and B<sub>2g</sub> channels that corroborate previously observed trends in the doping dependence of the B<sub>1g</sub> spectral weight depletion, and the relative immunity of the B<sub>2g</sub> spectra to changes in doping. In addition, in the most underdoped samples (x = 0.08 and x = 0.11) the B<sub>1g</sub> spectral weight at low energies is further depleted as the temperature is reduced from 300K to 50K. The spectra also show that this spectral weight appears to be transferred from the low-energies region to the higher energy (ω > 1500 cm<sup>-1</sup>) spectral region normally occupied by multi-magnon scattering. The observed temperature dependence of the scattered intensity in the B<sub>1g</sub> channel is found to be correlated with a depression of the scattering rate Γ(ω, T) in the B<sub>2g</sub> channel, similar to that derived from measurements of the infrared conductivity. The suppression of Γ(ω, T) is characterized by an energy scale ω<sub>Γ</sub>(x) ∼ 800 cm<sup>-1</sup> which is the same as the energy scale associated with the doping induced depletion of spectral weight. The results are consistent with those obtained using other techniques and suggest that the PG is magnetic in origin.

II. RESULTS AND DISCUSSION

A. Spectral Weight Depletion

In this paper all the as-recorded spectra have been divided by the Bose-Einstein factor to obtain the relevant Raman response functions. The B<sub>1g</sub> Raman response function of La214 is shown in Fig. 1 for different doping levels. It should be noted that all the spectra shown in

Fig. 1 are plotted on the same scale (after dividing the x = 0.22 spectrum by 5.5). This means, for example, that for the underdoped (x = 0.13) crystals, the integrated spectral weight in the B<sub>1g</sub> channel for 0 ≤ ω ≤ 800 cm<sup>-1</sup>, is about a factor of 6 times smaller than that for the overdoped (x = 0.22) crystal. Since the B<sub>1g</sub> spectra are dominated by scattering from excitations on regions of the FS located near the k<sub>x</sub> and k<sub>y</sub> axes, the results imply that underdoping leads to a significant depletion of low-energy spectral weight from these same regions of the FS [hereafter designated as the (π,0) regions]. We attribute this depletion of spectral weight to the presence of a PG which is approximately independent of temperature for doping levels x ≥ 0.13 and temperatures T ≤ 300K.

The doping dependence of the low energy spectral weight is summarized in figure 2. As is evident from Fig. 2 the strength of the depletion increases rapidly with decreasing doping. Alternatively one can say that the PG removes spectral weight from regions of the Fermi surface located near (π,0) and as we underdope the size of the affected regions increases significantly. The solid line in Fig. 2 was calculated using a simple tight binding band structure and assuming that the depleted arc length of the Fermi surface increases with decreasing doping. The qualitative agreement between experiment and calculation supports this effective fragmentation of
FIG. 2. The integrated low-energy ($0 \leq \omega \leq 800 \text{ cm}^{-1}$) $B_{1g}$ spectral weight plotted as a function of hole concentration at temperatures between 50K and 300K in La214. The data for $x = 0.17$ and 0.19 crystals are the same as those obtained at $T < T_c$ (Ref. 3) and have not been shown in Fig. 1 for clarity. The solid line is the result of a calculation (see text) while the dashed and dotted lines serve only as guides to the eye.

the Fermi surface. Furthermore, the additional reduction in spectral weight that occurs at low $T$, suggests that decreasing temperature leads to a further shrinking of the active Fermi surface area, as observed in a previous ARPES experiment [20].

As the doping level of La214 is decreased below optimum a broad peak appears in the $B_{1g}$ spectra at higher energies (Fig. 1). This peak is attributed to 2-magnon scattering and it grows in strength as the doping level is decreased. For $x \leq 0.11$ it becomes the dominant feature of the $B_{1g}$ spectra. Underdoping thus leads to an effective transfer of spectral weight from low energies to the higher frequency range occupied by the 2-magnon features. This observation is consistent with the suggestion [21] that the depletion of low energy spectral weight is associated with the presence of short range magnetic correlations. Such correlations are assumed to grow in strength as the doping level is decreased [4,22], thus removing an increasing amount of spectral weight (Fig. 2) from the FS and leading to an effective fragmentation of the FS. A more critical consideration of the spectra, however, indicates that the spectral weight gained in the 2-magnon frequency region is greater than the amount lost below 800 cm$^{-1}$, both as a function of doping and as a function of temperature. Although this observation may be somewhat surprising one must remember that the Raman intensity is proportional to a generalized density-density correlation function and thus one would not expect sum rules to apply rigorously [23]. In addition one must also note that the Raman continua extend to very high energies ($\sim 2\text{eV}$), which are outside the range of our spectrometer. It is possible, and perhaps even expected, that spectral weight will also be transferred from the region $\omega > 0.5\text{eV}$ to the 2-magnon region, as the doping level or temperature are reduced. Thus it is perhaps not too surprising that the observed enhancement in the 2-magnon region appears to exceed the low energy depletion.

In Raman experiments one-magnon scattering has not been observed but the magnetic excitations can be probed via the two-magnon scattering which is peaked [24–26] at $\omega_J \sim 3J$ in the undoped cuprates (Fig. 1). As the doping level is increased, and the AFM correlation length decreases, this feature broadens, weakens and softens in frequency. The peak frequencies associated with these 2-magnon features (which are designated $\omega_J$), have been estimated as shown in Fig. 1. The resultant values of $\omega_J$ are plotted as a function of doping in Fig. 3. If $\omega_J$ is assumed to be a measure of the strength of the short range AFM correlations the energy of the PG is then given by $E_g \sim \omega_J/3 \sim J(1-x/0.3)$ for $0 \leq x \leq 0.13$. Such a linear variation for $E_g$ was initially proposed by Loram et al. [11] from the results of specific heat measurements. We must emphasize however that the identification of the peak positions ($\omega_J$) is highly uncertain and thus the linear relation shown in Fig. 3 must be considered to be very tentative. It should also be noted that the spectra shown in Fig. 1 have not been corrected
for variations in the optical constants or for spectrometer response. Our spectrometer response decreases in the red and correction might push the peaks in Figs. 1(a-d), and hence the values of $\omega_J$ shown in Fig.1, to higher energies. Such corrections would not, however, alter the observed trend of $\omega_J$ to lower values as the doping level is increased.

**B. Temperature Dependence**

In the most underdoped samples the low-energy $B_{1g}$ spectral weight also decreases with decreasing temperature (Fig. 1). The integrated loss over this region ($\omega \leq 800/cm^{-1}$), as a function of temperature, is summarized in Fig. 2. It is clear that the temperature dependent depletion is smaller in magnitude, and appears to be superimposed on the doping induced depletion. The depletion induced by underdoping sets in at a temperature that is outside our range of observation (T > 300K). The results thus imply that there is an upper crossover temperature at which strong PG behavior appears and a lower crossover temperature $T^*$ below which a further contribution to the PG occurs. These results suggest the presence of two steps, or perhaps two mechanisms, associated with the depletion of low-energy spectral weight in underdoped crystals. The values obtained for $T^*$ from Fig. 1 for the $x = 0.08, 0.11, 0.13$ crystals are in qualitative agreement with estimates obtained for $T^*$ from FIR [8] and NMR [9] measurements in La214. Our results thus suggest that the PG is reflected in NMR $[10, 11]$ and FIR $[8, 10]$ measurements only in terms of the weaker temperature dependent depletion that occurs at $T^*$. The results also provide a reconciliation with specific heat measurements [11], in which no clear evidence for $T^*$ is obtained. Since these results are determined by an average of the spectral weight around the FS they will be most strongly influenced by the doping induced depletion.

The presence of short range AFM correlations can give rise to an anisotropic scattering rate $[2, 12]$ that results in the existence of “hot spots” near $(\pi, 0)$ and “cold spots” on regions of the FS near $(\pm \pi/2, \pm \pi/2)$. The depletion of the $B_{1g}$ spectra (Fig. 2) is consistent with the presence of “hot spots” and the properties of the “cold quasiparticles” should be reflected in the $B_{2g}$ spectra. These spectra (Fig. 4) are relatively independent of doping which is consistent with the presence of “cold spots” near $(\pm \pi/2, \pm \pi/2)$. This is somewhat puzzling however in that in infrared experiments evidence for the PG has been obtained from the frequency and temperature dependence of the scattering rate $1/\tau^*$ that is derived from measurements of the optical conductivity $[13, 14]$. In underdoped compounds the FIR response should be dominated by the “cold” quasiparticles $[15, 27]$. Thus this behavior should also be reflected in the $B_{2g}$ Raman response. However the $B_{2g}$ spectra we have obtained (Fig. 4) do not exhibit any obvious variation with either doping or temperature. The latter result is also in contrast to Raman data $[5]$ obtained from underdoped Bi2212, where the $B_{2g}$ intensity was found to decrease by about 10% when T was reduced from 250K to 20K.

**III. COMPARISON WITH INFRARED SPECTRA**

To investigate the temperature dependence of the $B_{2g}$ spectra more carefully we examine the variation of the scattering rate. We assume that the $B_{2g}$ response function can be described approximately by $[28]$

$$
\chi(\omega) \propto \frac{i/\tau^*}{\omega + i/\tau^*}.
$$

Qualitative agreement with the experimental spectra of Bi2212 is obtained $[24]$ if the scattering rate $1/\tau^*$ is assumed to vary with both frequency and temperature. To proceed we use an extended Drude model $[1]$ in which $1/\tau^*$ is given by:

$$
1/\tau^* \rightarrow \Gamma(\omega, T) - i\omega\chi(\omega, T),
$$

where $\Gamma(\omega, T)$ is the scattering rate, and the function $\omega\chi(\omega, T)$ is introduced to preserve causality $[1]$. Using this form of $1/\tau^*$, we arrive at an expression for the extended Drude response:
As shown in Figs. 5(a-c) the scattering rate $\Gamma(\omega,T)$ is suppressed (below a linear dependence) at 50K in the $x = 0.08$, 0.11, and 0.13 crystals. Such a depression could not be observed for $\Gamma(\omega,T)$ obtained at doping levels larger than $x = 0.13$ (see Fig. 3d, for example). The frequency $\omega_1$, below which $\Gamma(\omega,T)$ is depressed (indicated by arrows in Fig. 5), is approximately equals 800 cm$^{-1}$. Then, if the frequency $\omega_1$ can be taken \cite{3} as a measure of the PG energy, we can write $E_g(x) \simeq 800$ cm$^{-1}$. It is interesting to note that the energy scale of the depletion observed in the $B_{1g}$ channel is correlated with the suppression of $\Gamma(\omega,T)$ in the $B_{2g}$ channel. The results shown in Fig. 5 are consistent with the depression of low-energy scattering rates that have been obtained from FIR spectra \cite{10}. This observation is also consistent with the suggestion \cite{3,7,27} that FIR spectra are determined by the properties of $B_{2g}$ or “cold” quasiparticles.

IV. CONCLUSIONS

We have carried out Raman scattering investigation of the pseudogap in La$_{2-x}$Sr$_x$CuO$_4$ as a function of both doping and temperature. In underdoped compounds the presence of the pseudogap results in a depletion of spectral weight from regions of the Fermi surface located near $(\pi,0)$, which in turn is manifested by a loss of spectral weight in the $B_{1g}$ Raman spectra. Reducing the temperature from 300K to 50K leads to a further reduction of the low-energy spectral weight which is clearly observed only in the most underdoped samples. Furthermore, our spectra indicate that spectral weight lost from the low-energy region is transferred to higher frequencies, namely to the multimagnon region characterized by $\omega J \sim 3J$. We have also shown that the scattering rate depression that is used to characterize the pseudogap in infrared measurements can be obtained from the $B_{2g}$ Raman spectra which implies that the infrared spectra are dominated by contributions from “cold” quasiparticles. It is interesting to note that the Raman, infrared \cite{3}, specific heat \cite{11}, and ARPES measurements \cite{12} on La214 all suggest an energy $E_g \sim J$. Thus it appears that an experimental consensus for the pseudogap energy scale is emerging, at least in La214. Finally the energy scale and doping dependence that we have observed appear to be consistent with a pseudogap associated with short range antiferromagnetic correlations.

ACKNOWLEDGEMENTS

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