Evidence for magnetic pseudoscaling in overdoped $La_{2-x}Sr_xCuO_4$

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We report the results of electronic Raman-scattering experiments on an overdoped $La_{1.78}Sr_{0.22}CuO_4$ single crystal as a function of temperature. The scattering rate $\Gamma(\Omega \rightarrow 0,T)$ has been determined from the normal state B_{1g} spectra in the range $50 \leqslant T \leqslant 300$ K. $\Gamma(T)$ decreases linearly from 300 K to about 175 K and then undergoes a reduction with respect to the expected mean-field behavior. This trend suggests a crossover to a magnetic pseudoscaling regime at $T_{cr} \simeq 160$ K. The results are in good agreement with the prediction of the nearly antiferromagnetic Fermi-liquid model. There is no evidence of a pseudogap in the spectra obtained from this overdoped sample. [S0163-1829(98)50918-6]

It is now quite clear that the normal state properties of high temperature superconductors are very different from those of a Fermi liquid (FL). For example, in optimally doped materials the unusual nature of these properties is manifested in transport measurements[1–3] by a resistivity that varies linearly with temperature and in Raman experiments[4] by a featureless electronic continuum that extends to large energies. Recent studies[5–12] of underdoped compounds have revealed even more remarkable deviations from FL behavior. Many experiments have provided evidence for a strong quasiparticle renormalization, or depletion of spectral weight, that sets in at a temperature $T^* > T_c$. The results of most of these investigations have been interpreted in terms of the opening of a normal state pseudogap (PG), a term that is generally used to mean a large suppression of low energy spectral weight. Although a great deal of effort has been expended the physical origin of the PG remains unknown. Recent photoemission experiments[9,10] have found that the PG has the same symmetry as the superconducting gap. These results imply that the depression of spectral weight above T_c is associated with precursor pairing. For example, it has been proposed[13,14] that pair formation, without phase coherence, could occur at $T > T_c$ with phase coherence, and hence the transition to the superconducting state, being established at the lower temperature T_c. However, other experiments, such as the specific heat measurements of Loram *et al.*,[15] are consistent with a mechanism that competes with superconductivity for the available quasiparticles.

Another issue concerns the nature of the normal state excitations in the overdoped regime. Although there is experimental evidence to suggest that the overdoped compounds behave more like normal metals, there is considerable evidence to the contrary. Some workers[3,11,16] have found that the PG is still present well into the overdoped state. Transport measurements by Hwang *et al.*[11] on overdoped $La_{2-x}Sr_xCuO_4$ [La214(x)] suggest that T^* is much greater than T_c for $x \leqslant 0.22$. More recently, infrared reflectivity measurements[16] on La214(0.22) have been interpreted in terms of the onset of a PG at $T^* \approx 300$ K. These measurements suggest that the PG persists into the overdoped state and are consistent with a modified phase diagram proposed by Batlogg *et al.*[3] in which, for La214, T^* is significantly greater than T_c at optimum doping and becomes equal to T_c well into the overdoped region.

Recently Pines and co-workers,[17,18] after analyzing NMR and neutron-scattering experiments, have proposed a new phase diagram for the hole-doped cuprates that could provide an explanation for the apparently conflicting results described above. In their nearly antiferromagnetic Fermi liquid (NAFL) model a tendency to order antiferromagnetically competes for quasiparticles with a spin-fluctuation-mediated pairing mechanism. They have discussed their results in terms of the variation of the antiferromagnetic (AF) correlation length ξ with temperature and doping. This leads to the definition of a temperature T_{cr}, at which $\xi(T_{cr}) \cong 2a$, where a crossover occurs between what they define as a mean-field (MF) region ($T > T_{cr}$) to a region ($T^* < T \leqslant T_{cr}$) in which magnetic pseudoscaling (PS) prevails and ξ increases rapidly with decreasing temperature. At a lower temperature $T = T^*$ there is a crossover to the PG regime in which ξ is approximately constant. Both T_{cr} and T^*, and their difference, decrease with increasing doping and for La214 approach T_c well into the overdoped regime.[18]

To gain additional insight into the above issues concerning the normal state of overdoped cuprates, we have carried out electronic Raman-scattering experiments on an overdoped La214(0.22) single crystal. In many ways La214 is a prototype material for these studies in that it has a single CuO_2 layer in the unit cell and this enables one to avoid any effects that might be associated with interlayer coupling. In addition, the overdoped state of La214 is well characterized and can be reproducibly attained.[19–21] Finally, La214 has been the subject of many of the previous studies cited above and thus our results can be compared directly to these studies

and to the predictions of the model proposed by Pines and co-workers.[18] We have measured the low-energy B_{1g} and B_{2g} Raman continua as a function of temperature. From each B_{1g} spectrum we have obtained an estimate for the scattering rate $\Gamma(\Omega\rightarrow 0,T)$. The dependence of Γ(T) on temperature is consistent with the existence of a crossover from MF to a PS behavior at $T_{cr}\simeq 160$ K. We have not found any evidence for a crossover to a PG regime, however, which implies that $T^{*}\leqslant T_c$ for this overdoped sample. The results are in general agreement with the predictions of the NAFL model.

The high-quality La214(0.22) single crystal ($T_c=30$ K) used in this study was grown by a traveling solvent floating-zone method, as previously described.[19] The specimen, with dimensions of $4\times 2\times 0.5$ mm^3, were oriented using Laue x-ray diffraction patterns. The surfaces of the sample were polished with diamond paste and etched with a bromine-ethanol solution.[22] Raman spectra were obtained in a quasibackscattering geometry using the 514.5 nm line of Ar^+ laser, which was focused onto the sample with a cylindrical lens to provide an excitation level of about 10 W/cm^2. The temperature of the excited region of the sample was found to be about 11 K above ambient. This has been estimated from the intensity ratio of the Stokes and anti-Stokes spectra. The temperatures reported in this paper are the actual temperatures of the excited region of the sample.

The scattering geometries used in this paper are defined by specifying the polarizations of the incident and scattered light with respect to a set of axes, x(1,0,0) and y(0,1,0), which are chosen to lie along the Cu-O bonds in the CuO_2 planes, or with respect to x'(1,1,0) and y'(1,1,0), which are rotated 45° with respect to x and y. In all cases the incident and scattered light travels parallel to the z(0,0,1) axis. For nonresonant excitation the components of the Raman tensor are given approximately[23,24] by $\gamma_{ij}\propto\partial^2\epsilon(\mathbf{k})/\partial k_i\partial k_j$. The orthorhombic distortion is very small[19–21] for this overdoped crystal and thus tetragonal symmetry (D_{4h} point group) can be used for a discussion of the polarization dependence of the Raman experiments. In this case γ_{xy} must transform[23] as k_xk_y or the B_{2g} irreducible representation of D_{4h} and $\gamma_{x'y'}$ must transform as $(k_x^2-k_y^2)$ or B_{1g}. Thus $\gamma_{x'y'}$ has a maximum near the k_x or k_y axis and is zero along the diagonal directions, while γ_{xy} will have the complementary symmetry dependence in k space. In other words, the xy scattering geometry probes regions of the FS located near the diagonal directions and the $x'y'$ spectra arise from excitations located near the (1,0) axes. These symmetry considerations can be illustrated by using a tight-binding model to represent the band structure and then calculating[12] the angular dependence of the components of the Raman tensor. The result of such calculation carried out for $\gamma_{x'y'}$ is shown in the inset of Fig. 1.

In an attempt to gain information on the scattering processes that influence the B_{1g} channel, we have studied the temperature dependence of the B_{1g} spectra. Figure 1 shows the $x'y'$ Raman response of La214(0.22) obtained at temperatures between 50 K and 300 K. The Raman response functions for symmetry γ (χ''_γ) are obtained by dividing the measured intensity by the thermal factor $[1-\exp(-\hbar\Omega/k_BT)]^{-1}$. The dashed lines in the figure represent the slope of χ''_γ as $\Omega\rightarrow 0$. As shown in Fig. 1, the low-energy continua

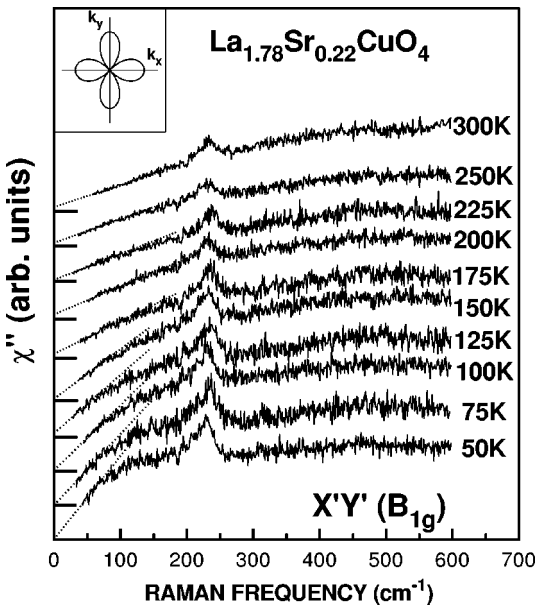

FIG. 1. The B_{1g} Raman response ($\chi''_{B_{1g}}$) of La214(0.22) obtained at temperatures between 50 K and 300 K. The dashed lines are the slopes of $\chi''_{B_{1g}}$ as $\Omega\rightarrow 0$. The spectra have been displaced for clarity and their zeros are indicated by ticks on the vertical axis. The inset shows a polar plot of the angular dependence of the B_{1g} component of the Raman tensor $\gamma_{x'y'}$.

indicate a redistribution which gradually decreases with increasing temperature and changes trend for $T>150$ K. As will be described below, this behavior might be associated with the variation of the AF correlation length ξ with temperature, which suggests a crossover from a MF to PS regime at $T_{cr}\simeq 160$ K. Furthermore, Figure 1 indicates an increase in the intensity of the Raman continuum near 500 cm^{-1}, once the temperature is decreased below 160 K. This behavior, as well as the higher frequency Raman response, will be discussed in detail in a forthcoming publication.

If both vertex corrections and the real part of the self-energy are neglected, an expression [Eq. (3) of Ref. 25] for the Raman response in the limit of $\Omega\rightarrow 0$ and $T\ll\mu$ can be written as

$$\chi''_\gamma(\Omega,T)=\frac{2\Omega}{N}\sum_{\mathbf{k}}\ \gamma^2(\mathbf{k})\frac{\Gamma^2(\mathbf{k},T)}{[(\epsilon(\mathbf{k})-\mu)^2+\Gamma^2(\mathbf{k},T)]^2}. \quad (1)$$

Here $\Gamma(\mathbf{k},T)$ is the momentum- and temperature-dependent scattering rate which is equal to the imaginary part of the self-energy evaluated on the FS, $\epsilon(\mathbf{k})$ is the band structure, μ is the chemical potential, and N is the number of sites. From Eq. (1) the slope of the low-energy Raman response $\chi''_\gamma(\Omega\rightarrow 0,T)$ is inversely proportional to $\Gamma(\mathbf{k},T)$ weighted by the Raman tensor $\gamma(\mathbf{k})$,

$$\left[\frac{\partial\chi''_\gamma(\Omega,T)}{\partial\Omega}\right]_{\Omega\rightarrow 0}\propto\Gamma_\gamma^{-1}(T)=\langle\gamma^2(\mathbf{k})/\Gamma(\mathbf{k},T)\rangle, \quad (2)$$

where we have replaced the $\mathbf{k}$ sum with an integral over an infinite band and an angular integral $\langle\cdots\rangle$ over the FS. Using Eq. (2) we can thus determine the B_{1g} scattering rate from the inverse slope of $\chi''_{B_{1g}}(\Omega\rightarrow 0,T)$ shown in Fig. 1.

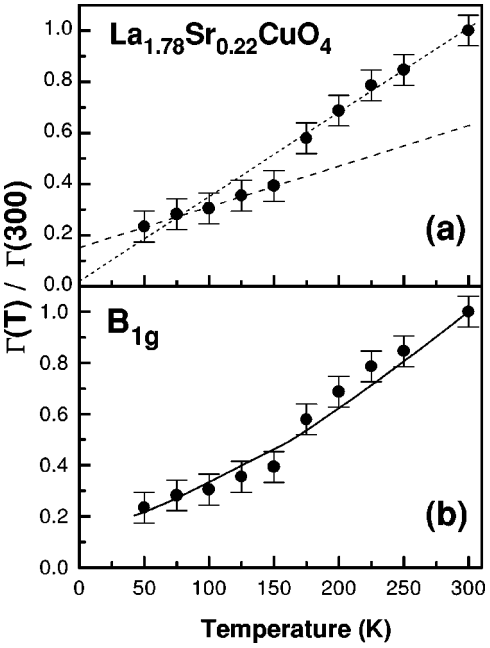

FIG. 2. (a) The normalized dc ($\Omega \to 0$) scattering rate $\Gamma(T)/\Gamma(300)$ in La214(0.22) obtained from the B_{1g} spectra. The dashed lines are least squares linear fits to MF and PS regimes. (b) Comparison of the normalized dc scattering rate with values (solid lines) calculated from the NAFL model.

The results of these determinations can be seen in Fig. 2 where they are plotted in normalized form $[\Gamma(T)/\Gamma(300)]$ as a function of temperature. The variation of the scattering rate with temperature, as shown in Fig. 2(a), clearly suggests the identification of two distinct regimes with a crossover at approximately 160 K. These results appear to be in qualitative agreement with the predictions of the NAFL model if we make the identification $T_{cr} \simeq 160$ K. This assignment is also in accord with the results of the determination of T_{cr} shown in Fig. 4 of Ref. 17 obtained from transport and susceptibility measurements of La214.

These considerations are quite persuasive but to make the comparison more quantitative the Raman spectra for NAFL can be calculated.[25] The results of the calculated scattering rates are compared to the measured values in Fig. 2(b) where the solid lines indicate the results of evaluating Eq. (2). Here we have used the electron-electron interaction[17]

$$V(\mathbf{q},\omega) = g^2 \frac{\alpha \xi^2}{1+(\mathbf{q}-\mathbf{Q})^2 \xi^2 - i\omega/\omega_{sf}}, \qquad (3)$$

to calculate the self-energy.[25] In Eq. (3), g is the coupling constant, and ω_{sf} and ξ are the phenomenological temperature-dependent spin-fluctuation energy scale and the correlation length, respectively, which can be determined via fits to magnetic response data.[17] These functional parameters obey certain relations, depending on different temperature and doping regimes. In the $z=1$ or PS regime, the spin correlations are strong enough to lead to changes from the classical MF theory $z=2$ regime. For each scaling regime, $\omega_{sf}\xi^z =$ const (i.e., temperature independent). As in Ref. 17 for the $z=1$ scaling regime ($T^* < T \leqslant T_{cr}$) we use $1/\xi = 0.23 + 2.25 \times 10^{-3} T$ [K] and further assume that $\omega_{sf}\xi = 72$ meV for La214(0.22) with $T_{cr} = 160$ K. In the absence

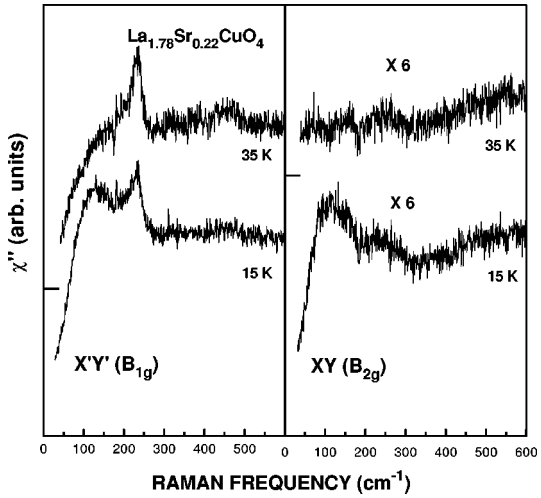

FIG. 3. The B_{1g} and B_{2g} Raman response functions (χ'') of La214(0.22) measured at 15 K and 35 K. The B_{1g} spectra are about 6 times stronger than the B_{2g} spectra.

of any prediction for how these parameters cross over from one scaling regime to another, we assume a direct crossover to $z=2$ behavior given by $\omega_{sf}^{z=2}(T)/t = \omega_{sf}^{z=1}(T_{cr})/t + 0.4k_B(T-T_{cr})/t$, with $t = 200$ meV the near neighbor hopping, such that for $z=2$ $\omega_{sf}\xi^2 = 122$ meV and the correlation length and spin-fluctuation energy smoothly cross over from $z=1$ to $z=2$ behavior at $T=T_{cr}$. In addition, we have used a tight-binding energy band with the $t'/t = 0.16$ and filling $\langle n \rangle = 0.8$ for both spins. Last, we add to Eq. (3) an isotropic impurity interaction $H_{imp} = \Sigma_{\mathbf{k},\mathbf{k}'} \Sigma_{i,\sigma} U e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{R}_i} c^\dagger_{\mathbf{k},\sigma} c_{\mathbf{k}',\sigma}$, where $\mathbf{R}_i$ denotes the position of the impurity labeled by i and U is the impurity potential. After averaging over the position of the impurities, this adds a momentum-independent term to the imaginary part of the self energy $\Gamma_{imp} = \pi n_i N_F |U|^2$, where n_i is the impurity concentration, and N_F is the density of states per spin at the Fermi level. To obtain the best fit to the data we have used $\Gamma_{imp} = 9.7$ cm^{-1}. The resultant agreement between theory and experiment is obviously quite satisfactory and makes a plausible case for the applicability of the NAFL. Further details are reported in Ref. 25.

According to Fig. 2(a) in both scattering regimes $\Gamma(T)$ varies linearly with temperature but with different slopes. In the mean-field region ($T > 160$ K) the electrons should be uncorrelated, and a linear extrapolation to $T=0$ should provide an estimate[26] for the dc impurity scattering rate $\Gamma_{MF}(0)$. A high temperature extrapolation is shown in Fig. 2(a) along with a linear extrapolation of the data for $T < 160$ K. As is evident the extrapolation from the pseudoscaling regime yields $\Gamma_{PS}(0) > \Gamma_{MF}(0)$. This increase in the estimate for the residual scattering rate might be associated with scattering by collective spin fluctuations which takes place in the PS regime. This suggestion is also consistent with the depletion of the low-energy B_{1g} spectrum at temperatures just above T_c (see Fig. 3).

To complete this discussion we note that the temperature dependence of the scattering rate does not provide any evidence for a crossover to a pseudogap regime. Furthermore, in underdoped crystals the presence of the PG is associated[12] with a significant depletion of spectral weight in the B_{1g}

channel as is also found in photoemission experiments.[9,10] This depletion resulted in the B_{1g} spectrum being much weaker than the B_{2g} spectrum. However, the B_{1g} spectrum of the overdoped crystal is very strong and in fact significantly more intense than the B_{2g} spectrum as is shown in Fig. 3. Finally, the presence of a PG tends to mask the transition to the superconducting state and hence there is no observable[12] superconductivity-induced renormalization in the B_{1g} channel. However, as is clear from Fig. 3, the B_{1g} spectrum undergoes a very strong renormalization when the sample is cooled below T_c. These considerations strongly suggest that the PG is absent in La214(0.22).

In summary, we have carried out polarized Raman measurements of the low-energy electronic continua of $La_{1.78}Sr_{0.22}CuO_4$ over a wide range of temperatures. From the low-frequency B_{1g} spectra we have estimated the temperature-dependent scattering rate $\Gamma(T)$ for quasiparticles on portions of the Fermi surface near $(\pm 1,0)$ and $(0,\pm 1)$ directions. The results appear to be in good agreement with the predictions of the nearly antiferromagnetic Fermi-liquid model and in particular a crossover from a mean-field to a pseudoscaling behavior at $T_{cr} \simeq 160$ K is suggested. However, we have not found any evidence for a crossover to a pseudogap regime. This would imply that $T^* \leqslant T_c$ for this overdoped sample.

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