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Raman investigation of the $YBa_2Cu_3O_7$ imaginary response function

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Results on the temperature dependence of the imaginary component of the response function $R''(\omega)$ in an untwinned $T_c \sim 90$ K Y-Ba-Cu-O crystal are presented. We find that the normal-state imaginary parts of the response functions for the distinct A_{1g} and B_{1g} symmetries exhibit behavior characteristic of a quasielastic-scattering mechanism, having a temperature- and frequency-dependent relaxation rate. Further, the zero-frequency resistivities for the two symmetries inferred from this quasielastic model closely resemble the measured resistivity for single crystals of this material. Finally, a low-energy linearity of the B_{1g} imaginary response function, accurately described in the normal state by this model in the limit $\omega < T$, is found to persist in the superconducting state but an analysis of the slope reveals a dramatic alteration at T_c.

It has been suggested that the background continuum evident in the Raman-scattering intensity of several of the high-T_c compounds behaves in a manner consistent with a temperature-dependent polarizability assumed by a recently proposed phenomenological *marginal*-Fermi-liquid model.[1–3] In this paper we present Raman data on the temperature dependence of the imaginary component of the $YBa_2Cu_3O_7$ (1:2:3) response function and conclude that the normal-state behavior is consistent with a model relaxational response function descriptive of scattering from fluctuations. While the damping term of this model response function indicates that the 1:2:3 superconductor lies at the limits of a Fermi-liquid description, only in the B_{1g} symmetry of the *tetragonal* representation do the imaginary-response-function data closely agree with the phenomenological theory. In the superconducting state we find that the B_{1g} component of the imaginary response function exhibits a significant modification. Specifically, the low-energy linearity ($\omega < 150$ cm^{-1}) present in the normal-state persists in the superconducting state but the slope begins to decrease below T_c and at 4 K has returned to approximately the 300-K value.

The untwinned single-phase Y-Ba-Cu-O $T_c \sim 90$-K crystals used in this study were grown according to a method described in detail elsewhere.[4] The crystals were mounted in a liquid-helium cryostat which allowed for precise control of the temperature down to approximately 4 K. The Raman data were taken at the various excitation energies of an argon-ion laser. As the broad continuum feature exhibited no significant resonance enhancement in this range of energies, we present only the result for the incident wavelength of 5145 Å. The light was dispersed by a three-stage scanning monochromator across a nitrogen-cooled photomultiplier tube, which collectively provided a resolution of 4 cm^{-1}. All measurements were taken at low power densities of approximately 10 W/cm^2 in order to minimize the local laser-heating effects, and the spectra have been corrected for the instrumental response. The temperatures were determined by taking a ratio of the Stokes to anti-Stokes spectral functions.

The fluctuation-dissipation theorem for Raman scattering requires that the Raman spectral density $S(\theta,\omega)$ obey

$$S(\theta,\omega)=[1+n(\omega)]R''(\Theta,\omega)\,, \qquad (1)$$

where $n(\omega)=[\exp(\hbar\omega/k_BT)-1]^{-1}$ is the temperature-dependent Bose factor, $R''(\Theta,\omega)$ is the imaginary component of an appropriate response function, and θ denotes the symmetry. For reasons previously discussed,[1] the symmetries of the Raman excitations for the 1:2:3 superconductor can be meaningfully described in the tetragonal representation.

The constraints that $R''(\Theta,\omega)$ must be an odd and continuous function indicate that on dividing out the thermal function from the spectral density the resulting spectra should tend toward zero intensity at low frequency. While the A_{1g} spectra (x^2+y^2 symmetry) of Fig. 1 exhibit the expected dropoff of intensity at low energy, the exact line shape of the low-energy background is obscured by the presence of the 115 and 150 cm^{-1} phonons. The 150-cm^{-1} Cu(2) vibrational mode is accurately described by a Lorentzian profile and can be straightforwardly extracted. The 115-cm^{-1} Ba-mode line shape, however, exhibits significant anisotropy previously identified as deriving from a coupling of this mode to the background continuum.[5] As the background likely reflects the electronic density of states $\rho(E)$, it is apparent from the sharp rise of the data near zero-energy shift that a low energy $\rho(E)$ is strongly energy dependent. This substantial energy dependence precludes a unique fitting of the low-energy phonon and continuum in these data.[6]

The spectral intensity in the 150–1000-cm^{-1} region of the 300-K $R''(A_{1g},\omega)$ spectra of Fig. 1 undergoes a significant alteration upon the decrease of the temperature to 100 K. The redistribution in this temperature range occurs in a manner consistent with a relaxational scattering function having a temperature- and frequency-dependent damping. In particular, the solid lines of Fig. 1 represent fits to

$$R''(\omega) \propto \omega\Gamma/(\omega^2+\Gamma^2)\,, \qquad (2)$$

$$\Gamma=[(\alpha\omega)^2+(\beta T)^2]^{1/2}\,, \qquad (3)$$

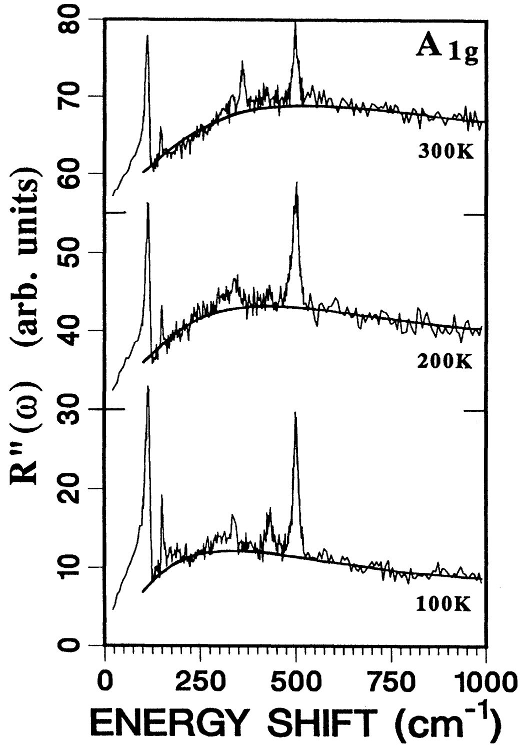

FIG. 1. Temperature dependence of the imaginary part of the A_{1g} response function. The solid lines represent fits to a simple relaxational response function and the short lines along the vertical border denote the zero-intensity level for the spectrum immediately above.

where α and β are free parameters of order unity and $k_B/\hbar$, respectively, the ratio α/β determines the high-energy dropoff of the function, T is the temperature, and the functional form indicates a simple relaxational response characteristic of quasielastic scattering from fluctuations. The values of the parameters used in these fits are shown in Table I.

While the fits seem adequately descriptive of the midenergy tail, the quasielastic model response for the 300-K spectra appears to fall off too slowly below 300 cm^{-1} to be appropriate. The disparity cannot be entirely due to the B_{1g} phonon at 330 cm^{-1} since fits to this phonon indicate that it makes no significant contribution to the scattering intensity below 300 cm^{-1} in the normal state.[7]

TABLE I. Values of the temperature and the damping coefficients for the fits of the quasielastic response function to the A_{1g} and B_{1g} data of Figs. 1 and 2.

α	β (cm^{-1}/K)	Temp. (K)	Symmetry
0.27	3.2	100	A_{1g}
0.27	2.1	200	A_{1g}
0.27	1.7	300	A_{1g}
0.75	2.2	100	B_{1g}
0.71	1.6	200	B_{1g}
0.65	1.5	300	B_{1g}

Rather, the inadequacy of the fit in this region may serve as a caveat to interpreting the model as a detailed description of the A_{1g} response. Instead, the functional form exposes a temperature-dependent trend of $R''(A_{1g},\omega)$ in the midenergy region involving both temperature- and frequency-dependent damping. Lending strength to this interpretation is the fact that the values of the damping term of Eq. (3) determined from the fits to the data reproduce a linear temperature-dependent resistivity quantitatively similar to direct resistivity measurements, as will be discussed below. Further, fits of the far-infrared conductivity to a Drude model involve a similar frequency- and temperature-dependent relaxation rate.[8]

The spectra of Fig. 2 were taken in the depolarized B_{1g} scattering geometry (x^2-y^2 symmetry). As the low-energy phonons are absent in this geometry for reasons detailed elsewhere,[9] an examination of the low-energy behavior of $R''(\Theta,\omega)$ is made possible. The solid lines represent fits to the quasielastic response function of Eqs. (2) and (3) indicating that the B_{1g} symmetry is accurately described by a model attributing the response to scattering from fluctuations. Distinguishing these fits from those of Fig. 1 is the fact that the fits to the B_{1g} spectra have roughly a three-times-larger ratio of the damping coefficients α/β. This indicates that the B_{1g} geometry has a more dominant frequency dependence on Γ than does the A_{1g} geometry. Investigations of several single crystals re-

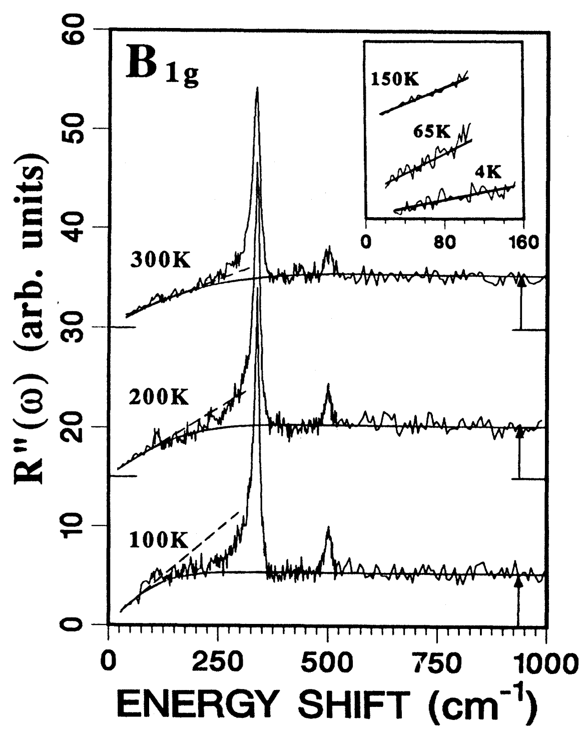

FIG. 2. Temperature dependence of the imaginary part of the B_{1g} response function. The solid lines represent fits to a simple relaxational response function and the vertical arrows mark the zero intensity level for the spectrum immediately above. The inset and the broken lines display the linearity of the low-energy region, and the inset data have been offset to make the linearity more apparent.

veal a scaling-factor variation of the continuum from sample to sample, but only a minor variation of these fitting parameters.

If the high-T_c superconductors do not lie in the dirty limit, approximate resistivities can be calculated from the damping terms of the quasielastic fits to the A_{1g} and B_{1g} spectra at zero frequency using the Drude result $\rho(T) \sim 4\pi\Gamma(T)/\omega_p^2$. The solid points of Fig. 3 show the resulting values for ρ assuming a value of $\omega_p = 1.4$ eV for the screened-plasma frequency.[10] The open squares are measured resistivity values for a representative $T_c \sim 90$-K Y-Ba-Cu-O twinned single crystal; however, the values can vary from sample to sample by as much as a factor of 3 larger.[11] The calculated values mimic the anomalous linearity of the resistivity indicating that the β parameters of the fits are only weakly temperature dependent. Further, the calculated values are within an order of magnitude of the measured values, suggesting that the dominant excitations responsible for the Raman continuum in the $T_c \sim 90$-K compound are provided by the current carriers of the resistivity measurements. Further support for this conclusion comes from Raman results on the oxygen-deficient compounds which reveal that the continuum weakens in a manner consistent with the decrease in availability of low-energy charge excitations on decreasing the carrier concentration into the insulating phase.[12]

In the appropriate limits, the B_{1g} quasielastic response can be seen to parallel the phenomenological hypothesis of Varma *et al.*[2] The broken lines at low energy for the three spectra seen in Fig. 2 result from least-squares fits to the data over an energy range from 20 cm^{-1} up to $\omega \sim 0.8$ T. The resulting values of the slopes for these low-energy fits are shown in Fig. 4, and the statistical deviation from linearity for the fits manifests itself in the error bars associated with the slopes at the different temperatures. Evident in Fig. 4 is the constancy of the low-energy normal-state behavior when the data are presented as a product of the slope and the temperature. This indicates that to within experimental resolution the normal-state B_{1g} imaginary part of the response function is roughly proportional to ω/T for energies $\omega < T$.

In the midenergy region (450 cm$^{-1} < \omega < 1000$

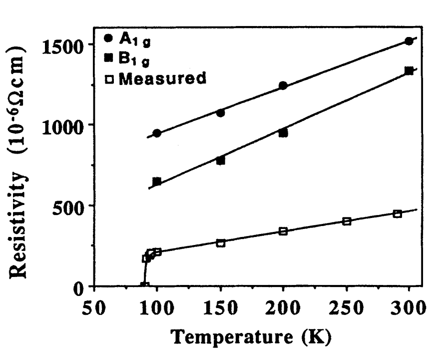

FIG. 3. The calculated and measured resistivities for single-crystal 90-K Y-Ba-Cu-O. The solid circles and squares represent the values determined from the A_{1g} and B_{1g} fits, respectively, and the open squares are the measured resistivity values from Ref. 11.

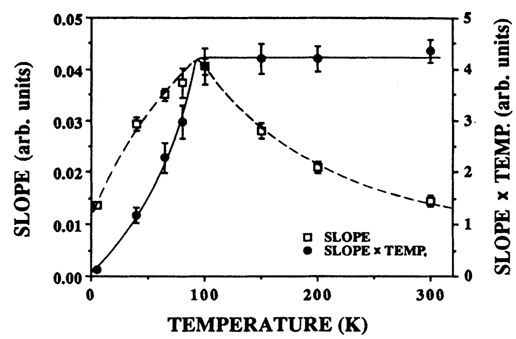

FIG. 4. Temperature dependence of the slope and the product of the slope and temperature for the low-energy linear component ($\omega < 150$ cm^{-1}) of the B_{1g} imaginary response function. The increased error bars for the slope×temp. data results from the uncertainty in the determination of the temperature. The lines serve as a guide to the eye.

cm^{-1}), the B_{1g} quasielastic fits are all equally displaced from the zero intensity value as the short vertical arrows in Fig. 2 indicate. This illustrates that to within experimental resolution, the midenergy component of $R''(B_{1g},\omega)$ is nearly a constant ($\sim S_0$) and temperature independent. Cumulatively, over the energy range 20–1000 cm^{-1} the imaginary part of the B_{1g} response function can be approximately described by

$$R'(B_{1g},\omega) \propto S_0\omega/T \text{ for } |\omega| < T , \tag{4}$$

$$R''(B_{1g},\omega) \propto S_0 \text{sgn}(\omega) \text{ for } |\omega| > T , \tag{5}$$

which is consistent with the polarizability proposed in the *marginal*-Fermi-liquid description.[2] This correspondence with the theory suggests that the B_{1g} Raman continuum shares its origin with that of numerous other normal-state properties of the high-T_c compounds.

While the A_{1g} model response has the same limiting form as Eqs. (4) and (5), the function is not found to fit the data at higher energies. Indeed, the quasielastic response of Eqs. (2) and (3) must have a limited applicability since the function has a divergent integral for large frequencies. Preliminary investigations reveal that for energies $\omega > 1500$ cm^{-1} the data in these geometries and in a geometry parallel to the c axis begin to deviate from the functional form of the model response.[13] This indicates that at high energies either the continuum response of Eq. (2) is made inadequate by the onset of additional mechanisms affecting the polarizability, or that the quasielastic response of Eq. (2) continues to be accurate but the damping of Eq. (3) undergoes significant alteration.

Although the quasielastic model is not descriptive of the superconducting state,[14] the low-energy region ($\omega < 150$ cm^{-1}) is accurately described by a linear term as the inset of Fig. 2 and previous results illustrate.[15] A comparison of the slope of this linear region below T_c to the slope of the linear segment of the model response in the limiting form of Eq. (4) then serves to elucidate the effect of the onset of superconductivity on the B_{1g} continuum. The comparison is made in Fig. 4. Below T_c, the value of the slope describing the low-energy component of $R''(B_{1g},\omega)$ decreases, and at 4 K is roughly 35% of the slope of the $T \sim T_c$ spectra. The suppression occurs over the entire temperature range below T_c and, relative to other superconducting properties illustrated through various Raman experiments, this effect is curiously gradual. It was found that the formation of the 350-cm^{-1} gap peak of the A_{1g} symmetry and 500-cm^{-1} gap peak of the B_{1g} symmetry occurred over a narrow (roughly 10-K) range below T_c.[16] Further, the strongly coupled 330-cm^{-1} vibrational mode was found to soften abruptly below T_c.[4,8,17] The disparity of these results may be an indication that these various experimental effects are not all directly linked to the opening of the superconducting gap.

If it is the gradual suppression of the slope which corresponds to the gap formation, then a model which attributes the continuum scattering to local fluctuations of a highly correlated electron liquid, would suggest that Fig. 4 details the shifting of the density of states below T_c.[1] Additionally, the presence of a nonzero linear scattering intensity at 4 K may indicate a persistence of these excitations well into the gapped state, a result consistent with previously reported Raman data,[4] and findings from various experimental techniques.[18] Further, the ratio of the slope at 4 K due to the residual excitations to the slope at 100 K due to the normal-state excitations may suggest that only 65% of the available carriers have condensed into the paired state at 4 K.

In summary, we have presented data that illustrate the temperature dependence of the imaginary response function $R''(\theta,\omega)$. We find that the trend of the normal-state response is accurately described by a simple relaxational response characteristic of scattering from fluctuations. Resistivities calculated from this model response suggest that these fluctuations derive from the current carriers of the resistivity measurements. In the appropriate limits, the B_{1g} response parallels the *marginal*-Fermi-liquid description indicating that the Raman continuum bears a close relationship to a variety of anomalous normal-state phenomena of the high-T_c cuprates. Finally, in the superconducting state, the slope of the linear low-energy region of $R''(B_{1g},\omega)$ undergoes a gradual suppression and at 4 K has returned to roughly its room-temperature value.

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[15]The results of Ref. 4 illustrate the linearity in the combined geometry $A_{1g}+B_{1g}$. The inset of Fig. 2 illustrates the low-energy linearity in the pure B_{1g} geometry.

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