Doping dependence of the pseudogap in the *ab* plane infrared spectra of $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_{8+\delta}$

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The *ab* plane optical spectrum of underdoped $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_{8+\delta}$ has been studied as a function of temperature. The gaplike suppression in scattering rate $1/\tau(\omega)$ observed below and above T_c is found to be related to the shoulder structure in reflectivity $R(\omega)$ and the broad peak or downward suppression from Drude-like shape in conductivity $\sigma_1(\omega)$. We conclude that the superconducting gap, which is visible in $\sigma_1(\omega)$, evolves smoothly into the pseudogap. The results obtained are consistent with recent electron tunneling and photoemission measurements. [S0163-1829(99)05201-7]

It is well recognized that the electronic properties of high-T_c cuprates are very different from those of conventional superconductors. One striking phenomenon is a gaplike feature below a certain temperature $T^* > T_c$ typically observed in underdoped $YBa_2Cu_3O_{7-\delta}$ (YBCO) by various techniques including NMR,[1] electronic specific heat,[2] *c*-axis optical conductivity,[3] and dc resistivity measurements.[4] This feature, commonly referred to as the pseudogap, removes only a fraction of states at the Fermi level as the material remains metallic. Recently, angle-resolved photoemission spectroscopy[5] (ARPES) and scanning tunneling spectroscopy[6] (STS) directly revealed the normal state pseudogap in underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) materials. The ARPES measurements indicate that the pseudogap has the same d_{x2-y2} symmetry and magnitude as the superconducting gap, pointing out its intimate relationship to superconductivity. A depletion of spectral weight in underdoped Bi2212 crystals above T_c was also observed from electronic Raman-scattering measurements,[7] which appears to be maximal in the B_{1g} scattering channel, analogous to the pseudogap indicated by ARPES measurements.

Optical spectroscopy is a fundamental technique in probing the electronic state of a superconductor. The search for a gap in the *ab*-plane infrared spectrum has been a task from the beginning of the discovery of high temperature superconductivity. The expected superconducting gap energy from the high-T_c value is large enough for detection in the far-infrared (FIR) region. However, because there are different approaches—one component or two-component—to analyze the conductivity spectrum, the question of whether excitations across a gap are observed in the *ab*-plane optical spectrum is still not resolved, even though an onset of absorption, which would correspond to 2Δ in ordinary superconductors, is seen between 300 and 600 cm^{-1} in many cuprates.[8] Recently, Puchkov *et al.*[9] have systematically analyzed the evolution of scattering rate $1/\tau(\omega,T)$ which was extracted from the *ab*-plane optical conductivity in a series of compounds from underdoped to overdoped. In underdoped region, they observed a gaplike depression in $1/\tau(\omega,T)$ opening up in the normal state. As the pseudogap is closely related to the superconducting gap, a careful examination of spectral change would shed light on the superconducting gap in *ab*-plane optical response.

In this paper we present the *ab*-plane optical spectra of underdoped $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_{8+\delta}$. A gaplike suppression in $1/\tau(\omega,T)$ is clearly observed in the normal state below a certain temperature, which deepens in the superconducting state. The onset of the suppression shifts to higher energy as compounds become more underdoped. We examine the reflectance and conductivity spectra closely and find that the suppression in scattering rate is related to the shoulder structure of the reflectance and to the suppression of conductivity. The results obtained from one-component analysis are in good agreement with STS and ARPES measurements. We conclude that the superconducting gap is visible in *ab*-plane optical conductivity spectrum.

Pure Bi2212 crystals were grown by the floating-zone method.[10] The Y-doped Bi2212 crystals were grown by usual melting method.[11] The pure crystal shows a narrow transition at 91 K, while the Y-doped crystals show broader transitions around 70 K for nominal Y concentration $x = 0.07$ and 30 K for $x = 0.11$, from resistivity measurements. Judging by their superconducting transitions, the pure sample is very close to optimal doping, while the Y-doped samples are in the underdoped regime. Freshly cleaved crystals were mounted on optically black cones, and the reflectance was measured from 50 to 9500 cm^{-1}, at temperatures above and below T_c, on a Bruker IFS 113*v* using an *in situ* overcoating technique.[12] The optical conductivities have been calculated from a Kramers-Kronig analysis of the reflectance, which requires extrapolations at high and low frequencies. At low frequency, the reflectance was extrapolated to zero frequency by assuming a Hagen-Rubens relation. Actually, the conductivity in the measured frequency range is found to be insensitive to different extrapolations at low ω. The reflectance has been extended to high frequency (40 eV) using the measurements of Terasaki *et al.*[13]

The measured *ab*-plane reflectance and the calculated real part of conductivity for the pure and

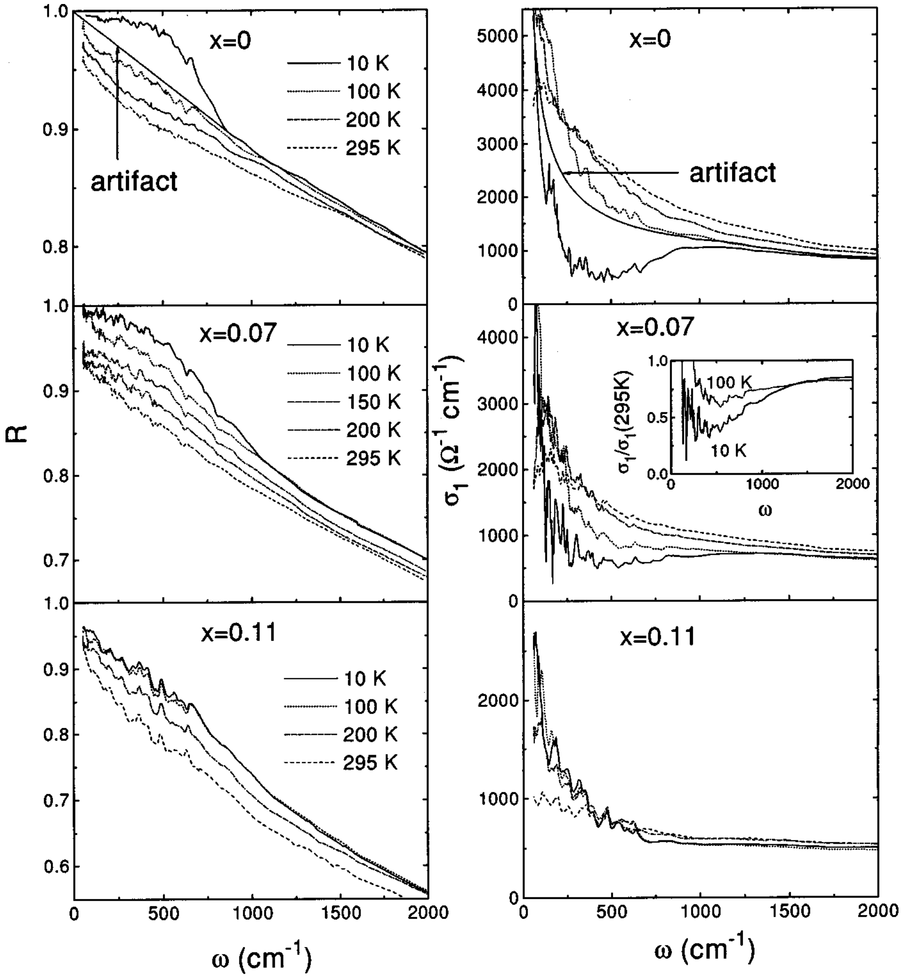

FIG. 1. The frequency dependence of the *ab*-plane reflectance (left panel) and conductivity (right panel) of $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_{8+\delta}$ single crystals with nominal Y concentration $x=0$, 0.07, and 0.11 at various temperatures. The curves indicated by artifacts are spectra with the shoulder in $R(\omega)$ removed by a straight line extrapolation. The inset is a plot of the ratio of the conductivity at 10 and 100 K to that at 295 K for the $x=0.07$ sample.

Y-doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ at several temperatures for $\omega < 2000\ \mathrm{cm}^{-1}$ are shown in the left and right panels of Fig. 1, respectively. For all three samples, the low-ω reflectance $R(\omega)$ and conductivity $\sigma_1(\omega)$ display a metallic T dependence, which is in agreement with the dc transport measurements.[10,11] A notable feature in $R(\omega)$ spectrum is a shoulder structure appearing at $\omega > 500\ \mathrm{cm}^{-1}$, which is the most prominent in the pure crystal at the lowest T (10 K). The reflectance is very close to unity and almost flat below 500 cm^{-1}, which is most likely due to the condensing of free carriers. This structure weakens as T increases, but is still visible in the reflectance curve at 100 K. A similar feature exists in the underdoped $x=0.07$ Y-doped sample. However, noticeable differences exist. First, the reflectance at 10 K is lower than that of pure sample and no longer flat below 500 cm^{-1}, suggesting an increased absorption by normal carriers. Second, the shoulder structure can be clearly observed in the normal state, for example, at 100, 150, and 200 K. Third, the shoulder becomes broad and extends to higher frequency; in the pure sample it ends at about 860 cm^{-1}, but extends to around 1050 cm^{-1} in $x=0.07$ sample. For heavily underdoped $x=0.11$ sample, the reflectance becomes much lower, which is attributed to a reduction in free carriers. There is only slight difference between the reflectance at 10 and 100 K, indicating that the proportion of condensed carriers is considerably reduced. In addition, phonon features become prominent at FIR frequencies. We think that this sample is close to the metal (superconductor)-insulator transition in the phase diagram. Nevertheless, a low frequency upturn at around 1100 cm^{-1}, similar to the shoulder structure, can be observed at all measured temperatures.

The conductivity spectrum $\sigma_1(\omega)$ is T-dependent in both FIR and midinfrared (MIR) frequency regions. Well below T_c, $\sigma_1(\omega)$ exhibits a broad maximum at frequency around 1130 cm^{-1} for the pure crystal. A further suppression is observed in $\sigma_1(\omega)$ below $\sim$860 cm^{-1} corresponding to the upturn in $R(\omega)$, which continues until $\sim$430 cm^{-1}; below this frequency a large amount of residual conductivity is seen and $\sigma_1(\omega)$ begins to increase with decreasing ω. For the underdoped $x=0.07$ sample, the broad maximum shifts to higher frequency, $\sim$1400 cm^{-1}, and the minimum appears at around 540 cm^{-1}. A weak dip at this frequency can also be seen at 100 K. This spectrum is obviously displaced downward from the usual Drude-like shape. Though a clear onset of suppression cannot be seen, it is estimated that the deviation starts at $\sim$1400 cm^{-1} as well. If we plot the ratio of the conductivity at low temperature to that at room temperature,

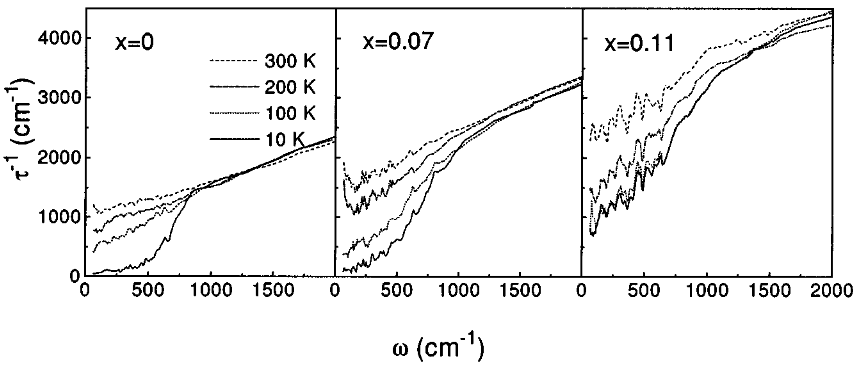

FIG. 2. The frequency dependent scattering rate for the three samples at different temperatures.

we find that the frequency dependent ratios for 10 and 100 K look very similar (see inset in right panel of Fig. 1). At higher temperature, e.g., at 200 and 295 K, the spectra show a continuous increase with decreasing ω below 2000 cm^{-1}. For the $x=0.11$ sample, the spectral weight is considerably reduced due to the reduction of free carriers. The spectrum is nearly ω-independent above 800 cm^{-1} and no clear suppression can be found in the conductivity spectrum.

It is well recognized that the strong electron correlation in high-T_c materials makes the simple Drude model with a constant scattering rate invalid. An alternative approach is to suppose a ω-dependent effective mass and scattering rate, $\sigma(\omega,T)=\omega_p^2/[4\pi(1/\tau(\omega)-i\omega(m^*/m))]$, which is normally referred to as the generalized Drude model.[8,9] The scattering rate, $1/\tau(\omega)$, and effective mass, m^*/m, can be obtained from the complex optical conductivity by $1/\tau(\omega)=(\omega_p^2/4\pi)\mathrm{Re}(1/\sigma(\omega))$ and $m^*/m=-(\omega_p^2/4\pi\omega)\mathrm{Im}(1/\sigma(\omega))$, where ω_p is the plasma frequency of charge carriers that can be estimated from the sum rule, $\omega_p^2/8=\int_0^\infty\sigma_1(\omega)d\omega$. An advantage of this model is that the frequency dependent parameters can be uniquely determined from the measured reflectance spectra. Plotted in Fig. 2, are frequency dependent scattering rates of the three samples at different temperatures. Here, plasma frequency values of $\omega_p=1.65\times10^4$ cm^{-1} for pure sample, 1.42×10^4 cm^{-1} for $x=0.07$, and 1.22×10^4 cm^{-1} for $x=0.11$, determined by summing the conductivity up to 1 eV,[14] have been used. As expected from the damping seen in the reflectance spectra, $1/\tau(\omega)$ increases as the sample becomes underdoped. In the pure sample, the scattering rate below T_c shows very sharp suppression around 860 cm^{-1}, which is believed to be due to the collapse of the free-carriers into the superconducting condensate. A slight suppression of $1/\tau(\omega)$ can be seen at 100 K in the normal state. At higher temperatures, $1/\tau(\omega)$ shows roughly linear ω dependent behavior. For the underdoped $x=0.07$ sample, the suppression of $1/\tau(\omega)$ can be seen not only at low T but also at temperatures much higher than T_c. Weak suppression is seen even at room temperature. A similar suppression of $1/\tau(\omega)$ is present in the heavily underdoped $x=0.11$ sample at all measured temperatures. Puchkov *et al.*[9] referred to such a gaplike suppression of $1/\tau(\omega)$ as the pseudogap in the *ab*-plane optical response. Here, we confirm such suppression in $1/\tau(\omega)$ above T_c in our underdoped Bi-based cuprates. We also note that $1/\tau(\omega)$ shows little T dependence at high-ω, which is also in agreement with the work of Puchkov *et al.*[9] However, the present work reveals additional features about the ''pseudogap'' in $1/\tau(\omega)$: (1) The suppression of $1/\tau(\omega)$ appears at the same frequency below and above T_c, being suggestive of the same amplitude for both superconducting gap and pseudogap. (2) Comparing the spectrum of $1/\tau(\omega)$ at 100 K with that at 10 K for underdoped $x=0.07$ sample, we can conclude that there is no abrupt change in $1/\tau(\omega)$ at T_c. The only difference caused by decreasing T is the depth of the suppression in $1/\tau(\omega)$. (3) The onset of suppression in $1/\tau(\omega)$ shifts to higher ω in underdoped samples, suggesting an increase of the characteristic energy for the pseudogap.

Because $1/\tau(\omega)$ is uniquely determined from the complex conductivity spectrum which is itself obtained by a Kramers-Kronig transformation from the measured reflectance spectrum, the gaplike suppression in $1/\tau(\omega)$ should manifest itself in the reflectance and conductivity spectra. A careful examination and comparison between these spectra indicate that the gaplike suppression in $1/\tau(\omega)$ is related to the shoulder structure in the reflectance. The onset of suppression in $1/\tau(\omega)$ corresponds closely to the onset of the upturn in the ω-dependent reflectance $R(\omega)$. The broad maximum in $\sigma_1(\omega)$, despite appearing at frequency higher than the onset of upturn in $R(\omega)$, is also related to the shoulder structure in $R(\omega)$. This can be confirmed by performing a Kramers-Kronig calculation on the reflectance with the shoulder structure removed—the broad maximum disappears. (See the reflectance and conductivity curves indicated by ''artifact'' in Fig. 1. The shoulder of the pure sample at 10 K is removed simply by a straight line extrapolation of $R(\omega)$ at $\omega=860$ cm^{-1} to unity at $\omega=0$.) Since the reduced absorption, or the shoulder, in $R(\omega)$ is considered to be due to the pairing of carriers, we believe that the suppression in conductivity is also due to this pairing. However, the suppression in $\sigma_1(\omega)$ is smeared or disappears in the underdoped samples or at high T. We think that this is mainly due to the d-wave nature of the pairing symmetry. Because the superconducting gap has a $d_{x^2-y^2}$ momentum dependent symmetry, a momentum-averaging measurement, such as infrared spectroscopy, would only detect a suppression feature rather than a full gap even at lowest temperature where the gap is fully formed. A theoretical calculation for a d-wave pairing[15] within a Hubbard picture shows that optical conductivity $\sigma_1(\omega)$ has a broad peak at 4Δ, where 2Δ is the maximal amplitude of d-wave gap. At low ω, there exists a residual Drude-like response due to scattering by nodal quasiparticles, which thus gives rise to a minimum for $\omega < 4\Delta$. In the presence of impurities, which should be closer to a real system, the states inside the gap can be easily filled, weakening the suppression below 4Δ. The calculated $\sigma_1(\omega)$ in their paper looks similar to our measured spectrum on the pure sample at the lowest T (10 K). Very recently, the evolution of the Fermi surface with temperature in underdoped Bi2212 has been mapped out by Norman *et al.* from ARPES.[5] The d-wave node below T_c becomes a gapless arc above T_c which expands with increasing T to form the full Fermi surface at T^*. Although a theoretical calculation of optical conductivity for such a gapless arc is lacking at this time, it is speculated that the expanding gapless arc would make the broad peak at 4Δ much weaker. This explains why the suppression becomes difficult to observe at high T. In addition, the density of uncondensed normal carriers increases relative to that of the paired carriers in underdoped samples. This also makes the broad maximum at the frequency 4Δ difficult to observe.

There has been long debate over whether a superconducting gap can be observed in the ab-plane $\sigma_1(\omega)$ spectrum, although a minimum was seen between 300 and 600 cm^{-1} in many cuprates. Schlesinger *et al.*[16] identified the characteristic energy scale of ~ 500 cm^{-1} observed in YBCO as a pair-excitation threshold 2Δ (superconducting gap) $(2\Delta/kT_c \sim 8)$. On the other hand, based on a two-component model, the observed absorption was explained as a tail of the T-independent MIR-absorption remaining after the condensation of the Drude part at low T.[8] Since we find that the spectral change in $\sigma_1(\omega)$ is related to that observed in $R(\omega)$ and $1/\tau(\omega)$, we identify the suppression in $\sigma_1(\omega)$ at low T as the formation of the superconducting gap. We also believe that the pseudogap manifests itself in the similar way, i.e., a similar suppression in conductivity spectrum. However, due to the reasons mentioned above, we only observe a downward deviation from the usual Drude-like spectrum above T_c in our underdoped sample. Actually, a much clearer suppression in ab-plane $\sigma_1(\omega)$ at $T > T_c$ was observed in underdoped YBCO samples previously.[17] The data in Ref. 17 also showed that the suppression was at the same energy for temperatures below and above T_c and shifted to slightly higher frequency for samples that were more underdoped. By reference to the theoretical calculation for a d-wave superconductor, which predicts a broad peak at frequency 4Δ in $\sigma_1(\omega)$,[15] we get the maximum gap $2\Delta \cong 565\ cm^{-1}$ for our pure Bi2212 $(2\Delta/kT_c \cong 8.8)$, but 700 cm^{-1} for underdoped $x = 0.07$ sample $(2\Delta/kT_c \cong 14.1)$. Note that in this theoretical calculation there is no particular structure at $\omega = 2\Delta$. Electron tunneling measurements[6] showed that the gap amplitude 2Δ increases from 42 meV (340 cm^{-1}) to 88 meV (710 cm^{-1}) for samples from very overdoped ($T_c = 56$ K) to underdoped ($T_c = 83$ K). Our results are in agreement with their work. ARPES (Ref. 5) also indicated that the gap amplitude increases in the underdoped sample, but gave smaller gap amplitudes.

There are considerable discrepancies over the origin of the pseudogap in cuprates. The pseudogap has been interpreted as a spin excitation gap—a pairing of spinons—based on the separation of spin and charge degrees which is supposed to occur in two-dimensional (2D) electron system,[18] or in a different opinion, to occur in locally 1D charge stripes formed by mobile holes.[19] The nearly antiferromagnetic (AM) Fermi liquid model suggested that the strong magnetic interaction peaked near AM wave vector would lead to the formation of a pseudogap due to a spin density wave precursor state, with a superconducting gap forming separately.[20] Additionally, it is proposed that incoherent pairs are performed above T_c, with phase coherences occur at T_c.[21] Our optical data show that the pseudogap is present in the ab-plane charge excitations, which would put a constraint on such theories. Furthermore, the present work and also the optical conductivity data on underdoped YBCO indicates that the pseudogap is closely related to the superconducting gap. The superconducting gap evolves smoothly into the pseudogap as T increases, which is most clearly evidenced from the scattering rate spectrum. These results suggest that the correlations that produce the pseudogap should be the very same ones that form the superconducting gap. Electron tunneling[6] and ARPES (Ref. 5) measurements on Bi2212 point in the same direction.

In conclusion, we have investigated the in-plane optical response of underdoped $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_{8+\delta}$. We found that the gaplike suppression in $1/\tau(\omega)$ observed below and above T_c is related to the shoulder structure in $R(\omega)$ and the broad peak or downward suppression from Drude-like shape in $\sigma_1(\omega)$. The superconducting gap amplitude has been determined from the conductivity spectrum, and becomes greater in underdoped samples. It should be pointed out that, although we can not rule out the two-component approach based on our optical data, the results obtained from one-component analysis are in good agreement with other measurements like STS and ARPES. The data suggest that the pseudogap has the same origin as the superconducting gap.

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