Anomalous microwave conductivity due to collective transport in the pseudogap state of cuprate superconductors

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The microwave surface impedance $Z_s = R_s + iX_s$ of $HgBa_2Ca_2Cu_3O_{8+\delta}$, $HgBa_2CuO_{4+\delta}$, $Tl_2Ba_2CuO_{6+\delta}$, and underdoped $YBa_2Cu_3O_{6.5}$ is found to be anomalous in that $R_s(T>T_c) \neq X_s(T>T_c)$ in the pseudogap state. This implies plasmonlike response and negative permittivities $\varepsilon'(\omega)<0$ at microwave frequencies indicating non-Fermi-liquid transport in the ab plane. The anomalous microwave response is shown to arise from a collective mode characterized by a plasma frequency $\omega_{pCM} \sim 0.1$ eV and extremely low damping $\Gamma_{CM} \sim 10^{-5}$–10^{-4} eV, distinctly different from those observed at optical frequencies.

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The "normal" state above T_c of the high-temperature cuprate superconductors is well known to be extremely abnormal. A wide variety of experimental techniques (photoemission, optical conductivity, NMR, tunneling, neutron scattering, infrared, Raman, etc.) (Ref. 1) have been applied to its study and suggest that there is a common phenomenology for all high-temperature superconductors: the existence of a partial gap or a pseudogap meaning the suppression of the low-energy density of states. An important issue is the nature of the pseudogap, several alternative theoretical models of this having been proposed, such as superconducting fluctuations[2] or islands,[3] competing order parameter,[4] and stripes.[5,6]

In this paper, we show that low-energy (microwave) measurements of the surface impedance $Z_s = R_s + iX_s$ on $HgBa_2Ca_2Cu_3O_{8+\delta}$ (Hg:1223), $HgBa_2CuO_{4+\delta}$ (Hg:1201), $Tl_2Ba_2CuO_{6+\delta}$ (Tl:2201), and underdoped $YBa_2Cu_3O_{6.5}$ reveal new features of transport in the pseudogap state. The measurements indicate a breakdown of the so-called Hagen-Rubens limit (where the measurement frequency $\omega \ll \Gamma$, the carrier relaxation or dissipation rate), indicating a plasmonlike response characterized by negative microwave dielectric permittivities $\varepsilon'(\omega)<0$, for currents in the ab plane. Such an anomalous conduction in the pseudogap state indicates non-Fermi-liquid (NFL) behavior rather than a single-particle (Fermi liquid) transport mechanism and that the microwave dynamics and the optical response are characterized by different energy scales. A model based upon a collective phason mode arising from the presence of charge fluctuations, such as from stripes or a density wave (DW), quantitatively explains the observed temperature dependence of experimental data.

Single crystals of Hg:1201 ($T_c = 94.4$ K), Hg:1223 ($T_c = 122$ K), Tl:2201 ($T_c = 91$K), and underdoped $YBa_2Cu_3O_{6.5}$ ($T_c = 60$ K) were prepared by appropriate methods for each material. The high quality of the crystals discussed here has been confirmed by a variety of other techniques.[7] The data reported here were confirmed with measurements on several samples of each material. The high-sensitivity microwave measurements of R_s and X_s were carried out in a Nb superconducting cavity resonant at 10 GHz in the TE_{011} mode with very high unloaded $Q \sim 10^8$.[8] Since $Z_s = R_s + iX_s = \sqrt{i\mu_0\omega/\tilde{\sigma}}$, from R_s and X_s it is possible to obtain σ_1 and σ_2, the real and imaginary parts of the conductivity, using $\tilde{\sigma} = \sigma_1 - i\sigma_2 = i\mu_0\omega/(R_s + iX_s)^2$. In all microwave measurements, $R_s(T)$ can be measured absolutely, while relative changes $\Delta X_s(T) \equiv X_s(T) - X_s(0)$ are typically measured. We obtain $X_s(0) = \mu_0\omega\lambda_{ab}(0)$ from estimates of the low-T penetration depth $\lambda_{ab}(0)$:130 nm (Hg:1223), 117 nm (Hg:1201), and 260 nm (YBCO6.5). It should be emphasized that because $X_s(\dot{T} > T_c) \gg X_s(0)$, the results discussed in this paper are not sensitive to $X_s(0)$ or $\lambda(0)$.

The temperature dependences of X_s and R_s for Hg:1223 when the microwave magnetic field $H_\omega \| c$ axis and of ΔX_s and R_s when $H_\omega \perp c$ are shown in Fig 1. In Fig. 1(a) $H_\omega \| c$ so that we are probing in-plane charge dynamics, while in Fig. 1(b), $H_\omega \perp c$ (i.e., $H_\omega \| ab$), the current is flowing in the ab and c directions. In this mixed case, the data are represented as ΔX_s because $\lambda_{ab+c}(0)$ and hence $X_s(0)$ cannot be easily estimated. At low $T \ll T_c$, $\lambda_{ab}(T)$ has a power-law depen-

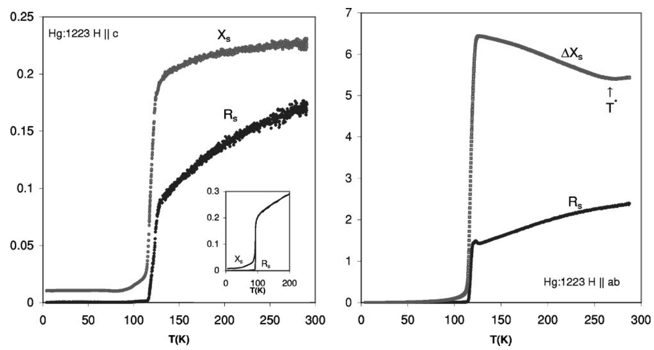

FIG. 1. (a) R_s and X_s vs T for $H_\omega \| c$ and (b) R_s and ΔX_s vs T for $H_\omega \perp c$ for $HgBa_2Ca_2Cu_3O_{8+\delta}$. The violation of the Hagen-Rubens limit in Hg:1223 is evident since $X_s \neq R_s$ for $T > T_c$ and $\Delta X_s(T_c) > \Delta R_s(T_c)$. Similar anomalous results are also observed in $Tl_2Ba_2CuO_{6+\delta}$, $HgBa_2CuO_{4+\delta}$, and underdoped $YBa_2Cu_3O_{6.50}$ (not shown). In contrast such a violation is not observed in optimally doped $YBa_2Cu_3O_{6.95}$ [inset to (a)].

dence on T, consistent with measurements on other cuprate superconductors.[9] Details of the superconducting state will be discussed separately.

Two principal features of the data for Hg:1223 of Fig. 1 are evident. (i) Above T_c the curves of R_s vs T and X_s vs T are not parallel, so that $R_s(T > T_c) \neq X_s(T > T_c)$. (ii) Furthermore, $\Delta X_s(T_c) > \Delta R_s(T_c)$, exactly opposite to that observed in conventional metals like Nb and Sn, and to within experimental accuracy, in optimally doped $YBa_2Cu_3O_{6.95}$ [see Fig. 1(a), inset] and $Bi_2Sr_2CaCu_2O_8$.

Essentially similar data were found for the other materials in this study: Hg:1201, Tl:2201, and $YBa_2Cu_3O_{6.5}$. The inequality $R_s(T > T_c) \neq X_s(T > T_c)$ for all four materials is evident from Fig. 2, where we present the data in terms of the anomaly $\mathcal{A} = X_s/R_s - 1$ vs T for both $H_\omega \| c$ [Fig. 2(a)] and as $\Delta X_s/R_s - 1$ vs T when $H_\omega \perp c$ [Fig. 2(b)]. The anomaly $\mathcal{A}$ is clearly finite (nonzero) for $T > T_c$ for both orientations. In optimally doped $YBa_2Cu_3O_{6.95}$ the anomaly $\mathcal{A}(T > T_c) = 0$ from the data of Fig. 1(a), inset.

The influence of the pseudogap temperature scale on the transport is clearly evident in Fig. 2 for Hg:1223 ($T^* = 270$ K) and Hg:1201 ($T^* = 260$ K).[10] The onset of the pseudogap greatly enhances the c-axis contribution, as is clearly seen in the data for Hg:1201 and Hg:1223 [Fig. 2(b)], although the onset can also be seen in the pure ab-plane data [Fig. 2(a)] albeit more gently. Thus our data are consistent with other findings that the c-axis pseudogap is different from the ab-plane pseudogap.[1]

For a conventional metal, the electromagnetic response can be expressed in terms of the dynamic conductivity written as $\tilde{\sigma}(\omega) \equiv \sigma_1 - i\sigma_2 = \sigma_{n0}/(1 + i\omega\tau) = \omega_p^2 \varepsilon_0/(\Gamma + i\omega)$, where ω_p is the plasma frequency, $\Gamma = \tau^{-1}$ is the relaxation or dissipation rate, and σ_{n0} is the zero-frequency (dc) conductivity. In typical metals like Al, $\omega_p \sim 15$ eV and $\Gamma \sim 0.1$ eV, a negative dielectric constant is observed at optical frequencies ($\sim 10^{13}$Hz, 0.1 eV) where $\omega \sim \Gamma$, and $\sigma_2/\sigma_1 \sim 1$, since $\tilde{\varepsilon} = 1 - \omega_p^2/\omega(\omega - i\Gamma)$. On the other hand, microwave frequency ($\sim 10^{10}$Hz, 10^{-4} eV) experiments are in the Hagen-Rubens limit $\omega \ll \Gamma$, $\sigma_2/\sigma_1 = \omega/\Gamma \ll 1$, implying $\tilde{\sigma} = \sigma_{n0} = \omega_p^2 \varepsilon_0/\Gamma$, and the conventional Ohm's law applies. In the Hagen-Rubens limit, $R_s = X_s = \sqrt{\mu_0\omega/2\sigma_n} = \mu_0\omega\delta_n/2$, where the skin depth $\delta_n = (2/\mu_0\omega\sigma_n)^{1/2}$. Clearly then our finding that $\mathcal{A}(T > T_c) = X_s/R_s - 1 \neq 0$ shows that these materials violate the Hagen-Rubens condition in the pseudogap state.[11]

The violation of the Hagen-Rubens limit immediately implies a finite value of the imaginary part $\sigma_2(T > T_c)$, since $\sigma_2 = \mu_0\omega(X_s^2 - R_s^2)/(R_s^2 + X_s^2)^2$ and a corresponding negative microwave dielectric permittivity $\varepsilon'(T > T_c) = -\sigma_2(T > T_c)/\omega\varepsilon_0$, for the *nonsuperconducting* state above T_c for these materials. $\sigma_2(T)$ is shown in Fig. 3. In Hg:1223 it achieves rather large values $\sim 10^6$ $(\Omega\,\mathrm{m})^{-1}$ and decreases with increasing temperature. The corresponding dielectric constant $\epsilon = \sigma_2/\omega\varepsilon_0 = -2 \times 10^6$ is large and negative. Tl:2201 also violates the Hagen-Rubens limit, with values of $\sigma_2 \sim 10^5$ $(\Omega\,\mathrm{m})^{-1}$ leading to $\epsilon = -2 \times 10^5$. Essentially similar results have been found for Tl:2201 in other microwave measurements.[12] In underdoped $YBa_2Cu_3O_{6.5}$ the corresponding $\sigma_2(T > T_c)$ values $\sim 10^4$ $(\Omega\,\mathrm{m})^{-1}$ are significantly lower. Thus the violation of the Hagen-Rubens limit is less severe (although unambiguous) and is consistent with the trend that in optimum-doped $YBa_2Cu_3O_{6.95}$, $\Delta X_s(T_c) < \Delta R_s(T_c)$ and $R_s(T > T_c) = X_s(T > T_c)$ so that $\sigma_2(T > T_c) \sim 0 (\ll \sigma_1)$ within experimental error.

The above conclusions concerning finite $\sigma_2(T > T_c) \sim \sigma_1(T > T_c)$ are directly a consequence of the data and not

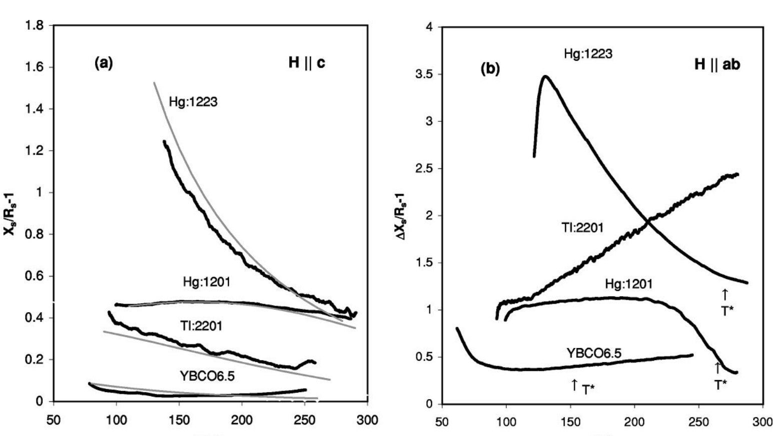

FIG. 2. (a) Experimental data (dark lines) for the anomaly $\mathcal{A} = X_s/R_s - 1$ vs T and the model (light lines) when $H_\omega \| c$. (b) $\Delta X_s/R_s - 1$ vs T when $H_\omega \perp c$. The arrows indicate the pseudogap temperature T^*.

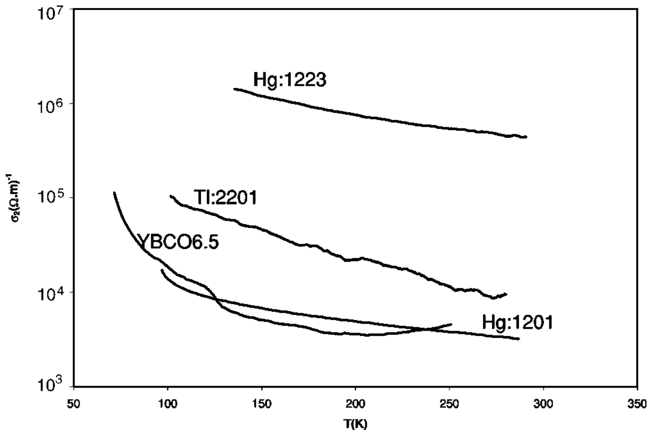

FIG. 3. $\sigma_2(T>T_c)$ for Hg:1201, Hg:1223, Tl:2201, and YBCO6.5.

obtained from any modeling of the dynamics. In the framework of a Drude relaxation model $\tilde{\sigma}(\omega)\equiv\sigma_1-i\sigma_2=\sigma_{CM0}/(1+i\omega_{CM}\tau_{CM})=\omega_{pCM}^2\varepsilon_0/(\Gamma_{CM}+i\omega)$ valid at microwave frequencies, we can obtain ω_{pCM} and Γ_{CM} from the $\sigma_1(10\ \text{GHz},T)$ and $\sigma_2(10\ \text{GHz},T)$ data. The resulting values of $\omega_{pCM}\sim0.1$ eV are significantly lower than indicated by optical spectra. More striking are the extremely low dissipation or scattering rates $\Gamma_{CM}\sim10^{-5}$–10^{-4} eV. These low values of Γ_{CM} are to be expected from the finite σ_2 since $\sigma_2/\sigma_1=\omega/\Gamma_{CM}\sim0.1$–1. Similar small values of Γ and also ω_p are observed in the heavy fermion materials UPt_3 ($\omega_p/2\pi\sim0.3$ eV, $\Gamma\sim6\times10^{-5}$ eV) (Ref. 13) and the conducting polymer polypyrrole (0.007 eV, 1.2×10^{-4} eV) (Ref. 14) from microwave measurements. The temperature dependences of $\omega_{pCM}(T)$ and $\Gamma_{CM}(T)$ are shown in Fig. 4. In Hg:1201 and Hg:1223, the temperature dependences appear to be tied to the pseudogap temperatures although no consistent trend is apparent.

Optical experiments show a Drude peak at much higher frequencies than microwaves, leading to the parameters $\omega_{p,opt}/2\pi\sim2$ eV and $\Gamma_{opt}\sim0.1$ eV. These magnitudes also correspond to ARPES measurements of the quasiparticle scattering rate. It is clear that these high-energy experiments are not able to observe the very low dissipation rates reported in this experiment, since $\omega\gg\omega_{pCM},\Gamma_{CM}$ for them, and further the collective mode observed in this work has small spectral weight $\int\sigma(\omega)d\omega=\pi\omega_{pCM}^2\varepsilon_0/2$ and $\omega_{pCM}\ll\omega_{p,opt}$. Thus our results clearly show a disparity in energy scales between the microwave and optical frequency transport.

Using the Drude form $\omega_{pCM}^2=ne^2/m^*\varepsilon_0$, we can extract the effective mass m^*. For n we use conventional estimates of 0.2 holes per plane, leading to $n\sim10^{27}/m^3$. The resulting effective masses then are somewhat large, $m^*\sim300$–400 for Hg:1223, 100–200 for Tl:2201,~100 for $YBa_2Cu_3O_{6.5}$, and 8000–23 000 for Hg:1201 in the temperature range of the data. These large masses are comparable with those observed in one-dimensional (1D) charge density waves (CDW's).[15] The simultaneous enhancement of $\tau_{CM}(=\Gamma_{CM}^{-1})$ and m^* leaves σ_1 nearly unchanged, so the microwave anomaly consists of a large value of $\sigma_2(T>T_c)$. It should be noted that earlier analysis of microwave scattering rates in Bi:2212 and $YBa_2Cu_3O_{6.95}$ (Ref. 16) assumed an effective mass $m^*=m_e$ (no mass enhancement) so that the Γ deduced from the microwave conductivity for $T>T_c$ in those cases is much larger than those deduced here. Massive carriers with $m^*\sim10^3$ have been deduced from microwave measurements in nonsuperconducting La_2CuO_4.[17]

The microwave results are suggestive of a phason mode of a CDW, whose electrodynamic response can be represented as $\tilde{\sigma}_{CM}(\omega)=\sigma_{CM0}/(1-\omega_{pin}^2/\omega^2+i\omega\tau_{CM})$. In the unpinned case $\omega\gg\omega_{pin}(\rightarrow0)$, the response reduces to the Drude form $\tilde{\sigma}_{CM}(\omega)=\sigma_0/(1+i\omega\tau_{CM})$ used above. If the phason has a finite pinning frequency, the model cannot explain the dc conductivity, which is actually enhanced below the pseudogap transition. To approximately describe this residual conductivity, we introduce a second Drude component, which is unaffected by the pseudogap transition. For simplicity, we assume the same marginal Fermi liquid form for the unrenormalized scattering of both components: $\sigma=\hat{\sigma}_0/(\tau_{MFL}^{-1}+i\omega m^*)+r_c\hat{\sigma}_0/(\tau_{MFL}^{-1}+i\omega)$, with r_c the ratio of the ungapped to gapped contributions, $\hat{\sigma}_0=\omega_p^2\varepsilon_0$, and $\tau_{MFL}^{-1}=\sqrt{\omega^2+\pi^2(T^2+T_0^2)}$, T_0 providing a low-temperature cutoff. The CDW effective mass enhancement is $m^*/m=1+\zeta$, with $\zeta=a\Delta^2(T)=\zeta_0(1-T/T^*)$, where $\Delta(T)$ is the gap, assumed to have a BCS form.[18]

Figure 2 shows the resulting calculated variation with T of the measured anomaly $\mathcal{A}=(X_s-R_s)/R_s=\gamma-1+\sqrt{1+\gamma^2}$ compared with the experimental data for the case $H_\omega\parallel c$

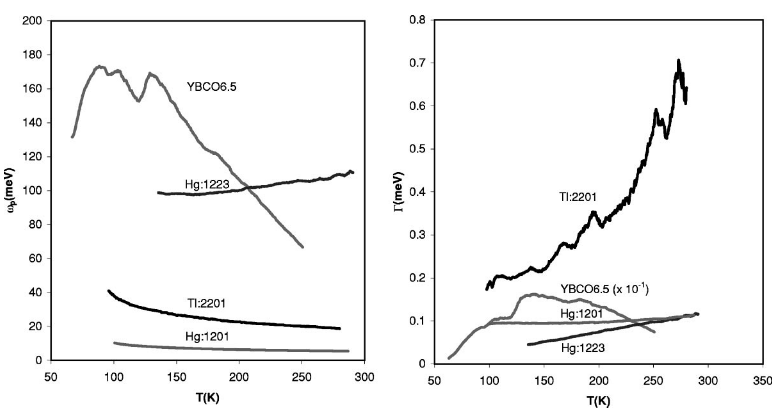

FIG. 4. (a) Plasma frequency ω_{pCM} and (b) dissipation rate Γ_{CM} in meV vs T for Hg:1201, Hg:1223, Tl:2201, and YBCO6.5, for $H_\omega\|c$.

where $\gamma=\sigma_2/\sigma_1$. Parameters are $(m^*, T^*, T_0/T^*, r_c) = (1000, 400\ \mathrm{K}, 0.3, .85)$ for Hg:1201, $(400, 450\ \mathrm{K}, 0, 0)$ for Hg:1223, $(500, 400\ \mathrm{K}, 0.5, 0.9)$ for Tl:2201, and $(150, 400\ \mathrm{K}, 0.3, 6)$ for $\mathrm{YBCO}_{6.5}$. The model reproduces the temperature dependence of $(X_s-R_s)/R_s$ found experimentally, decreasing at higher temperatures. The calculated T^* is an onset temperature T^*_{on}, while the experiment measures a crossover T^*_{cr}, where X_s/R_s changes most rapidly. We have compared only the case $H_\omega \| c$ for the pure ab-plane currents, since the mixed case $H_\omega \perp c$ requires an an additional c-axis contribution and is the subject of future work. We note that the microwave data do not find any clear indications for pinning of the phason mode (i.e., σ_2 is positive).

We have thus demonstrated that a collective mode approach is capable of explaining the anomalous microwave data, while requiring a high-frequency component for explaining the optical data. Since pair fluctuations persist only for a few K above T_c, the phenomena discussed here must be associated with pseudogap dynamics, rather than superconducting dynamics.[19]

In conclusion, we have presented microwave experiments that unambiguously reveal entirely novel transport properties of the nonsuperconducting or pseudogap state of several high-temperature superconductors. The pseudogap state has been probed at microwave time scales in several of these materials. The results show that the low-frequency transport is likely to be collective in nature, consistent with earlier suggestions of NFL above T_c (Ref. 20) and characterized by extremely low damping distinctly different from optical transport parameters. The results are quantitatively explainable in terms of a collective phason mode. Such a phason mode response can arise from a DW order parameter[4] or also from stripe fluctuations, which have CDW-like dynamics.[21] The implications of these results both for the pseudogap state, as well as the pseudogap-superconductor transition, are intriguing and of considerable importance.

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