Use of Kramers-Kronig relations to extract the conductivity of high-T_c superconductors from optical data

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In principle the conductivity of the cuprate superconductors can be obtained from reflectivity measurements using the Kramers-Kronig-transform technique. However, at low temperatures and for frequencies below $\sim 300\ cm^{-1}$ the reflectivities of materials such as $YBa_2Cu_3O_7$ are close to unity. Uncertainty in the precise signal level corresponding to unity reflectivity and a lack of knowledge of the reflectivity below the lowest measured frequency cause this method to become unreliable. To address this problem we have used a bolometric technique and a resonant technique to obtain accurate submillimeter and microwave data for the residual losses in epitaxial thin films of $YBa_2Cu_3O_7$ at low temperatures. The Kramers-Kronig analysis of our data is in good agreement with results from fitting our data to simple weakly coupled grain and two-fluid models for the *a*-*b* plane conductivity. However, below 450 cm^{-1} it is in disagreement with some published results of other workers obtained from Kramers-Kronig analysis of reflectivity data. To understand this discrepancy we analyze how the conductivity determined by the Kramers-Kronig-transform technique depends on some commonly used low-frequency extrapolations of reflectivity data.

Knowledge of the electronical conductivity $\sigma(\omega)$ is valuable for understanding the dynamics of the superconducting state of the oxide superconductors such as $YBa_2Cu_3O_7$ (YBCO). In practice, $\sigma(\omega)$ is often determined from Kramers-Kronig (KK) transform of reflectivity data. In principle, the KK technique requires knowledge of the reflectivity at all frequencies. In practical cases the reflectivity is measured over some finite-frequency interval and extended to zero and infinite frequencies by suitable extrapolations. The KK technique can be particularly troublesome in the low-frequency, low-temperature region where the measured reflectivity R approaches unity since neither the precise signal level corresponding to unity reflectivity nor the functional form of the reflectivity is known.

There are two common low-frequency extrapolations used for materials such as YBCO. One technique[1] is to use the Hagen-Rubens relation for the absorptivity $A=(1-R)=\alpha\omega^{1/2}$, which assumes the behavior of a normal metal at frequencies for which $\omega\tau \ll 1$. The value of α is chosen so that the extrapolation smoothly joins the reflectivity data at some frequency ω_0 below which the measured reflectivity is judged not to be reliable. While there is a clear justification for using this extrapolation to describe the reflectivity of a metal in the normal state, using it to describe the low-frequency, low-temperature loss of a superconductor such as YBCO is less justified. A second technique[2] for suppressing noise in the reflectivity is to simply set $A=0$ ($R=1$) below ω_0. This results in $\sigma_1=0$ below the same ω_0. While a value of $\sigma_1=0$ is consistent with our expectations for the behavior of low-T_c superconductors at low temperatures and for frequencies well below the gap, there is little experimental evidence from the cuprate superconductors to support this dependence.

Although unnecessary confusion has arisen because some authors do not adequately describe their extrapolations, the effect of uncertainties in the low-temperature reflectivity on the conductivity determined from the KK transform is widely suspected. For example, it has been briefly discussed by Orenstein *et al.*[2] Progress in determining accurate values of $\sigma(\omega)$ really depends on the availability of better low-frequency data.

In order to improve the experimental situation we have developed a technique to directly measure the low-temperature absorptivity of epitaxial *a*-*b* plane films in the frequency range between microwave loss and infrared reflectivity measurements.[3] In our measurement the thin film is used as the absorber in a composite bolometric detector[4] used to detect the signal from a Fourier-transform spectrometer with a Hg arc source. The data are normalized by comparison to reference detectors with normal-metal absorbers whose absorptivities can be calculated. This approach has several advantages over reflectivity measurements when the sample reflectivities being studied are close to unity. Uncertainties associated with the precise determination of unity reflectivity are minimized. In addition, sources of error which tend to multiply the measured spectra (e.g., drift, sample placement errors, standing waves) which can dominate values of $1-R(\omega)$ deduced from reflectivity measurements are reduced. We obtain absolute absorptivities below 100 cm^{-1} which have uncertainties as small as $\sim 5\%$ of the absorptivity.[3]

The residual losses for the films used in this study were also measured near 10 GHz and 4 K using microwave cavity techniques. For our best films, these are among the lowest reported in the literature. The microwave loss can be connected to the submillimeter data by a line which varies as frequency squared, that is $A=\beta\omega^2$, for each of our films. This interpolation agrees with microwave measurements by others[5] in the range from 10 to ~ 100 GHz (0.3–30 cm^{-1}). We use the same frequency squared dependence to extrapolate our data to zero frequency and use the KK-transform technique to determine the frequency-dependent conductivity, $\sigma(\omega)$, from our loss data.

In contrast to the low-frequency extrapolation, the choice of a high-frequency extrapolation can contribute an additive constant to $\sigma(\omega)$ for frequencies far below the extrapolation. We extend our absorptivity data to higher frequencies in two different ways. We have smoothly grafted our data, which ends at 700 cm^{-1}, onto reflectivity data for a good quality epitaxial film[6] which extends to 5 eV (40 000 cm^{-1}). This reflectivity is roughly constant near 5 eV, with $R \sim 0.08$. We then extrapolate the data to infinite frequency by assuming that the reflectivity remains constant above 5 eV. This is similar to the technique used by Orenstein *et al.*[2] We have also grafted our loss data onto the data of Tajima *et al.*,[7] which extends to 35 eV. Above this frequency we have extrapolated the reflectivity measured by Tajima *et al.* by an ω^{-4} dependence, which is the free-electron asymptotic limit. This is similar to the technique used by Schlesinger *et al.*[1] Despite the difference between these extrapolations, the conductivities determined by the KK transform for both of these high-frequency extrapolations are nearly identical below 2000 cm^{-1}.

We now use the infrared and microwave results for the high-quality epitaxial *c*-axis film *B* from Ref. 3 to illustrate the effects of various low-frequency extrapolations. The solid curve in Fig. 1 gives $\sigma_1(\omega)$ for film *B* from the KK method with the ω^2 low-frequency extrapolation. This curve rises slightly from its value at zero frequency to peak at ~ 50 cm^{-1}, and then drops to a minimum at 450 cm^{-1} above which there is a well-known onset of absorptivity which has been observed by many workers.[8] This is our best estimate of the correct $\sigma_1(\omega)$ for film *B*. The dotted lines in Fig. 1(a) correspond to the KK transform of the same absorptivity data which gives the solid line, but are obtained by setting $A=0$ below the cutoff frequencies $\omega_0 = 30$, 70, and 140 cm^{-1}. One effect of this artificial low-frequency extrapolation is to introduce a prominent absorptivity edge precisely at ω_0, below which $\sigma_1 = 0$. This artificial suppression of σ_1 at low temperature can introduce errors into so-called "missing area" calculations,[9] where the superconducting penetration depth is determined from the difference in oscillator strength between the low- and high-temperature values of $\sigma_1(\omega)$. The error will tend to decrease such estimates of the penetration depth, but only inversely as the square root of the area difference. The $A=0$ extrapolation also leads to values for $\sigma_2(\omega)$ which, in the vicinity of ω_0, deviate strongly from the result obtained from our ω^2 extrapolation.

We have also analyzed the effects of the $A=\alpha\omega^{1/2}$ low-frequency extrapolation of the KK transform. The dotted lines in Fig. 1(b) correspond to σ_1 obtained from the KK transforms of the absorptivity data analyzed with the absorptivity extrapolated as $\alpha\omega^{1/2}$ below the cutoff frequencies $\omega_0 = 30$, 70, and 140 cm^{-1}. As in Fig. 1(a), the solid line in Fig. 1(b) comes from our ω^2 low-frequency extrapolation. All of the curves for σ_1 obtained with the $A=\alpha\omega^{1/2}$ extrapolations closely approximate the solid curve above the respective values of ω_0. Below the cutoff frequencies, however, they diverge from the solid curve and provide an upper bound to the solid curve. Results for σ_2 obtained from the $A=\alpha\omega^{1/2}$ extrapolations are all within $\sim 20\%$ of the result obtained from the ω^2 extrapolation for frequencies above ~ 50 cm^{-1}. Below ~ 50 cm^{-1} the curves diverge.

The results of our KK analysis of our absorptivity data for films *A*–*E* of Ref. 3 are shown as solid lines of $\sigma_1(\omega)$ in Figs. 1 and 2. These results are qualitatively similar for the low absorptivity films *A*, *B*, and *D*.

The discussion presented above suggests that the small peak in $\sigma_1(\omega)$ at ~ 50 cm^{-1} observed for film *B* could arise from an error in the ω^2 interpolation between submillimeter and microwave data. A larger peak is seen in film *D* where $\sigma_{max} \sim 15\,000$ $(\Omega\,cm)^{-1}$ and $\sigma_{dc} \sim 12\,000$ $(\Omega\,cm)^{-1}$ In fact, by varying the parameter $\beta = A/\omega^2$ by $\pm 20\%$ we were able to either remove or introduce sharp peaks and valleys in $\sigma_1(\omega)$ near ~ 50 cm^{-1}. Even though the ω^2 interpolation between submillimeter and microwave data represents a significant improvement in our understanding of the frequency-dependent losses, it is not sufficiently accurate to eliminate all errors from the KK analysis. Consequently, the KK analysis presented here must be considered unreli-

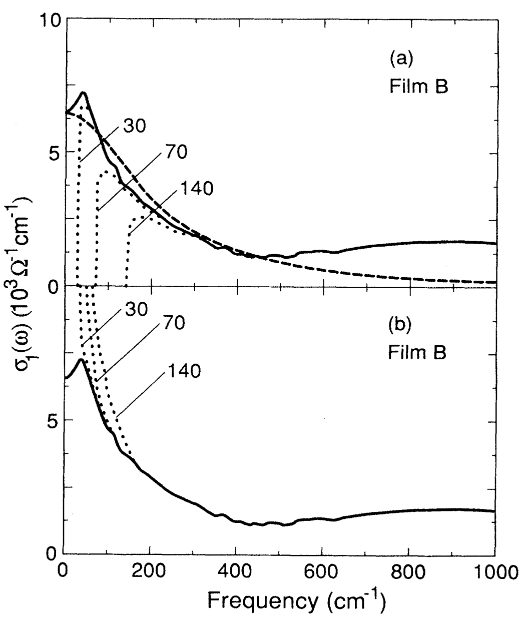

FIG. 1. Conductivity $\sigma_1(\omega)$ determined from a KK transform of our direct absorptivity data extrapolated to low frequencies as $A=\beta\omega^2$ (solid line) for the low absorptivity epitaxial *c*-axis film *B*. This result is compared with curves for $\sigma_1(\omega)$ obtained from the KK method by extending the absorptivity data with (a) an $A=0$ extrapolation and (b) an $A=\alpha\omega^{1/2}$ extrapolation below 30, 70, and 140 cm^{-1} (dotted lines). Also shown in (a) is $\sigma_1(\omega)$ obtained from a best fit to our absorptivity data using the weakly coupled grain model (dashed line).

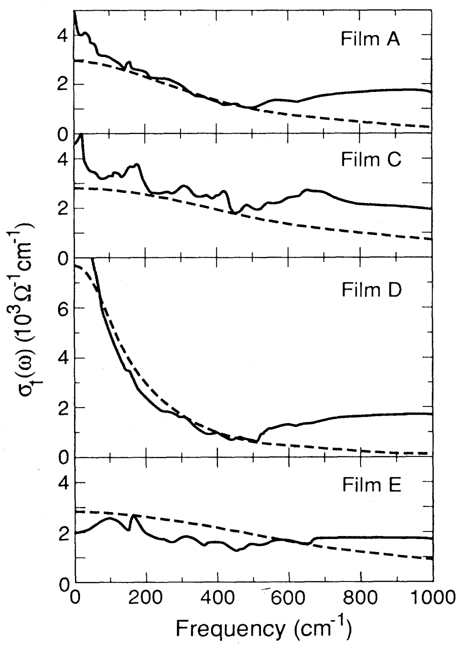

FIG. 2. Conductivity $\sigma_1(\omega)$ determined from a KK transform of our direct absorptivity data extrapolated to low frequencies as $A=\beta\omega^2$ (solid lines) compared with the best fit to our absorptivity data using the weakly coupled grain model (dashed lines) for films A, C, D, and E. Samples A and D are high-quality c-axis films. Samples C and E are of lower quality as discussed in Ref. 3.

able below ~ 50 cm^{-1}.

The absorptivities measured below 700 cm^{-1} are larger for films C and E than for films A, B, and D. Films C and E are thought to be of significantly lower quality. Film E was intentionally sputtered in an oxygen-deficient atmosphere to produce large residual microwave loss. The solid curves of $\sigma_1(\omega)$ deduced by the KK method for films C and E plotted in Fig. 2 are significantly different from the results shown for the high-quality films. The appearance of phonon structure, for example, demonstrates either the existence of crystallites whose c axis is not perpendicular to the sample plane or of considerable surface roughness.

In order to interpret the measured losses, we have used the weakly coupled grain model[10,11] to fit the submillimeter and microwave absorptivity data measured for each of our films. This model treats the a-b plane of polycrystalline high-T_c films as a network of grains made up of ideal BCS superconductors weakly coupled by resistively shunted Josephson junctions. This phenomenological, three-parameter model is valid for frequencies below the superconducting gap, and is not expected to describe phonon absorption or the additional absorption mechanism which is present above ~ 450 cm^{-1}. When the parameter corresponding to the superconducting penetration depth within the grains, λ_g, is constrained by the penetration depth obtained from muon spin rotation (μSR) measurements,[3,12] we obtain a good fit to the absorptivity data. Our use of the μSR data to constrain λ_g in the weakly coupled grain model is consistent with the interpretation of the μSR measurement for inhomogeneous materials.[13] The fit obtained in this way can also be interpreted as a two-fluid model in which the penetration depth λ_{tf} varies from 200 nm in our films with lowest loss to 520 nm in our highest loss films. From this fit we obtain the curves for $\sigma_1(\omega)$ shown in Figs. 1(a) and 2 (dashed lines). These curves are in good agreement with the KK-transform results for the low loss films A, B, and D in the range from ~ 50 to ~ 450 cm^{-1}. In addition, values for the superconducting penetration depth λ_{tf} determined from the KK transform, which are obtained from the low-frequency slope of $\sigma_2(\omega)$, are consistent with the results of our model fitting for the low loss films. In contrast, despite the fact that the weakly coupled grain model is able to fit the loss data for all five films below ~ 450 cm^{-1}, the agreement between the best fits and the KK transforms is not good for the high absorptivity films C and E. For film C, the weakly coupled grain conductivity does fit well to the minima between the phonon modes. This result may be expected as the grain model is fitted to the measured absorptivity only at points between the modes. Thus, the effects of the phonons are not included in the grain model. The two curves of $\sigma_1(\omega)$ for film E appear to be displaced by an additive constant. There is a special problem with the KK analysis of the absorptivity data for the loss film E which is thought to be deficient in oxygen. The measured absorptivity for film E does not intersect the high-frequency extrapolations described above which are appropriate for high-quality films. Various plausible linear interpolations have been used which cause 20% variations in the values of $\sigma_1(\omega)$ above 700 cm^{-1}. If the high-frequency absorptivity of film E is very different from that assumed, the results for $\sigma_1(\omega)$ could be shifted by an additive constant throughout the frequency range shown.

The results of our KK transforms of data for high-quality films are in agreement with the results of other workers both in magnitude and overall shape for frequencies above ~ 400 cm^{-1}.[1,2,14–17] In particular, we observe the well-known conductivity onset at 450 cm^{-1}. Below 400 cm^{-1} we observe that the conductivity decreases slowly from a peak at $\omega=0$. The values of $\sigma_1(0)$ which we have found from our fits to the grain model range from 3000 to 8000 $(\Omega\,\text{cm})^{-1}$ for the YBCO samples A–E. This result is consistent with two recent reflectivity experiments,[16,17] one of which was extended with a measurement of the microwave loss at 87 GHz.[17] It is interesting to note that our results are also consistent above ~ 100 cm^{-1} with the a-axis conductivity determined from the directly measured absorptivity of a detwinned single crystal of $YBa_2Cu_3O_7$.[18] However, below 400 cm^{-1} our results are in disagreement with the low temperature, low frequency conductivity determined from KK transforms of other reflectivity data,[2,14,15] from a combined transmissivity and reflectivity measurement,[19] and from a direct absorptivity measurement on twinned single crystals.[20] Uncertainties introduced into the KK transform by arbitrary low-frequency extrapolations may be responsible for these discrepancies.

To summarize, we have determined $\sigma(\omega)$ from a KK transform of absorptivity data which was directly measured between 30 and 700 cm^{-1} at 2 K and near 10 GHz (0.3 cm^{-1}) at 4 K. We have used two different high-frequency extrapolations and an $A=\beta\omega^2$ extrapolation below 30 cm^{-1} to extend the loss data to zero frequency. The low-frequency extrapolation is consistent with measurements on the same films near 10 GHz. The conductivities σ obtained from this analysis are in good agreement with results obtained from fitting our absorptivity data to the weakly coupled grain model when the superconducting penetration depth in the grains is constrained by the μSR result. We have compared our results to those obtained using less physical extrapolations found in the literature. The $A=\alpha\omega^{1/2}$ extrapolation provides an upper limit to the low frequency $\sigma_1(\omega)$ obtained from a truncated data set. In contrast, the $A=0$ extrapolation is more dangerous because it introduces an artificial gap-like feature in $\sigma_1(\omega)$ at the cutoff frequency ω_0 which is not present in the more realistic data set.

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