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Microwave study of the high-T_c superconductor $La_{1.8}Sr_{0.2}CuO_4$

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High-T_c superconductivity is investigated by a microwave-cavity perturbation technique in the compound $La_{1.8}Sr_{0.2}CuO_4$. Both the microwave loss and frequency shift show a rapid decrease at 39 K, the onset of the superconducting transition. A significant microwave absorption is also observed well below the transition. The data are used to determine the sample resistivity in the normal state and characterize the superconducting transition in an applied magnetic field.

Since the discovery of high-temperature superconductivity in metallic oxides, a number of subsequent magnetic and transport measurements[1,2] have been performed on these compounds to identify the conduction mechanism and the nature of the superconducting state. A dc resistivity measurement is mainly concerned with the normal state above the transition, while magnetization gives information on the superconducting state below the transition. ac measurements are of great interest because they can give information about both the superconducting and the normal states. Microwave[3] and far-infrared spectroscopy[4] are still useful even if a superconducting path is established and dc measurements cease to give information. They also give access to the real and imaginary parts of the complex ac conductivity. In the present work we have employed a microwave cavity perturbation technique to examine the superconducting transition in the bulk superconductor $La_{1.8}Sr_{0.2}CuO_4$. A similar technique has been employed in the past to characterize the inhomogeneous superconducting transition of granular aluminium films.[3] Both the microwave loss and the cavity frequency shift are generally utilized to obtain information about the superconducting state.

The experiments were conducted according to a standard cavity perturbation technique.[5] A cavity operating at 16.8 GHz was used in a TE_{102} transmission mode. The resonance curves were obtained on a digitizer by sweeping the rf frequency over the cavity resonance. A minicomputer automatically measured the resonance frequency f and the quality factor Q. This technique consists of measuring the frequency shift $\Delta f/f$ and the variation of the quality factor $\Delta(1/2Q)$ when the sample is inserted in the cavity at the maximum electric field location. Both parameters were registered with precision as the temperature was varied over the superconducting transition; to obtain power-independent data, the microwave power was maintained at minimum.

Polycrystalline samples of $La_{1.8}Sr_{0.2}CuO_4$ were prepared from La_2O_3, CuO, and $SrCO_3$ powders (all of 99.999% purity) mixed together and heated in air in an alumina crucible. The sample was held at 900 °C for 8 h and then cooled in the furnace. It was ground and reacted in air at 1100 °C for 18 h. This latter procedure was repeated once in air and a final time under an oxygen atmosphere. The sample was pressed into pellets and annealed for another 6 h. X-ray measurements indicated that this material was a single phase having the expected K_2NiF_4 structure. Resistivity and magnetic-susceptibility measurements were performed on the pressed pellets. The sample showed superconductivity with a transition midpoint of 35.7 K and a transition width of 7 K; the room-temperature dc resistivity value was approximately 25 000 $\mu\Omega$ cm and the value just above the transition $\rho_{41} \approx 8070$ $\mu\Omega$ cm.

The sintered samples were cut from a pressed pellet and had typical dimensions $5 \times 0.5 \times 0.5$ mm^3, a geometry which may be approximated with sufficient accuracy to an elongated ellipsoid. The cavity and sample were then placed in a double calorimeter head filled with helium exchange gas to ensure temperature stability and subsequently immersed in a liquid-helium bath. For magnetic field measurements up to 7 T, the setup was placed in a Nb-Ti superconducting coil. The temperature is monitored and stabilized with a Lakeshore controller with either a Si diode sensor or a $SrTiO_3$ capacitor for the magnetic-field data. An overall precision of 0.2 K was obtained for the 4–300 K temperature range.

The microwave loss $\Delta(1/2Q)$ and the frequency shift $\Delta f/f$ data obtained at 16.8 GHz for $4 < T < 300$ K are presented in Fig. 1. The frequency shift is constant for $T > 39$ K, the value being given by a geometrical factor according to the usual cavity perturbation theory applied to a highly conducting material. In the same temperature range the microwave loss increases almost linearly with the temperature, indicating that the resistivity is effectively increasing toward room temperature. Both parameters decrease rapidly with temperature at 39 K and practically saturate below 32 K. The rapid decrease of both parameters demonstrates that the sample becomes superconducting in agreement with dc resistivity and susceptibility measurements. This loss is easily understood because the electric field is rapidly expelled from the sample when it

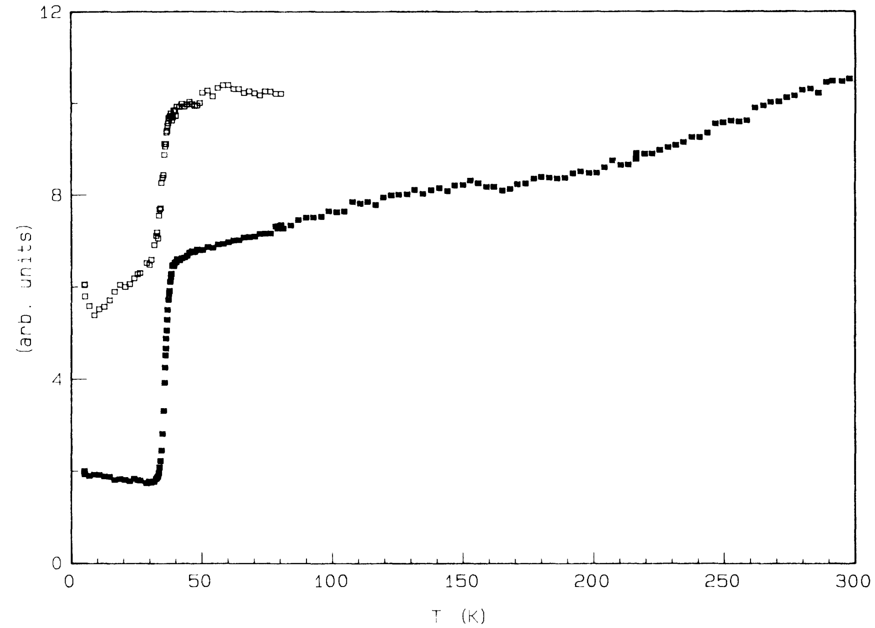

FIG. 1. Microwave loss $\Delta(1/2Q)$ (■) and frequency shift $\Delta f/f$ (□) at 16.8 GHz as a function of temperature.

becomes superconducting, a situation which yields a dramatic decrease in the microwave absorption. The other interesting feature in these data is the relatively high loss obtained below the superconducting transition; even after a superconducting path is established and dc measurements cease to give information, a microwave loss is still measurable and it can be used to characterize the superconducting sample below the transition.

As no clear mechanism has yet come out to explain the significant loss observed below the superconducting temperature ($T < 39$ K), these data will not be treated here. In the normal state ($T > 39$ K), the microwave field does not penetrate the sample entirely as its thickness is greater than the skin-depth in the normal state. Thus, only a limited portion of the sample (thin shell) is indeed absorbing. In this case, the skin-depth approximation equations[6] must be used to compute the sample resistivity. The resulting resistivity is shown in Fig. 2; just above the transi-

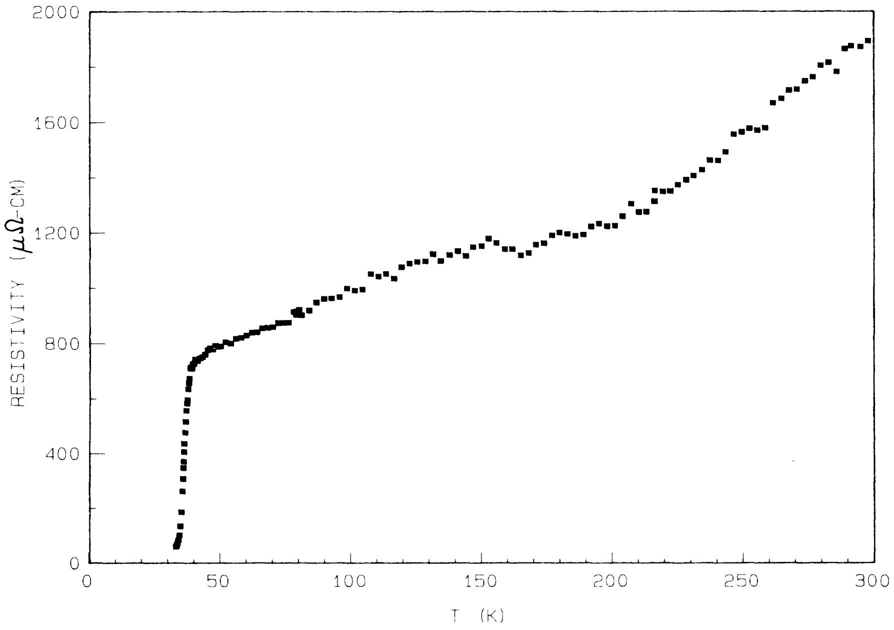

FIG. 2. Microwave resistivity at 16.8 GHz as a function of temperature.

tion it has the value $\rho_{39} \sim 700\ \mu\Omega$ cm and it increases almost linearly to a room-temperature value of ca. $\rho \sim 1900\ \mu\Omega$ cm. These values of the resistivity are smaller by a factor around 12 than the ones obtained by the dc technique. This is not really surprising as the microwave measurement is not sensitive to the intergrain resistance; our values are thus more close to the intrinsic resistivity value of the material. A change in the slope of the resistivity near 200 K is probably due to the same orthorhombic distortion observed by neutron-diffraction in $La_{1.85}Sr_{0.15}CuO_4$ in this temperature range.[7] These values are smaller than the ones obtained from a four-probe configuration dc measurement. This is, however, consistent with the nature of the sintered sample, which should offer more resistance to a dc current. The onset of the transition on the microwave loss and frequency shift is at 39 K with a midpoint at 35.8 K and the (10% to 90%) width is 3.95 K.

When an external magnetic field is applied, the microwave loss and frequency shift data are only modified for $T < 39$ K. The effects are weak on the shift but important on the loss; these last ones are shown in Fig. 3 for $20 < T < 50$ K. The observed magnetic field dependence is typical of what is generally found in the superconducting state. The rapid decrease observed below 39 K is shifted toward lower temperatures with increasing magnetic field while the width of the transition is increasing. The saturation value of the loss at lower temperatures is enhanced by the application of the field. According to these data, a linear relation is found between the applied field and the midpoint transition temperature; by extrapolation a fairly large upper critical field $H_{c2}(T=0) \sim 38$ T is deduced with a temperature dependence $-dH_{2c}/dT|_{T_c} = 1.53$ T/K. This last value is in agreement with the one obtained by Orlando *et al.*,[8] and Batlogg *et al.*[9] on a sample of the same composition. As it is often observed in superconductors, the width of the transition is an increasing function of magnetic field (0.76 K/T).

Another interesting feature of our microwave experiment is the fairly large microwave loss observed below the transition, 26% of the loss measured just above the transition. When the magnetic field is applied to the sample, this value jumps rapidly to 30% for $H < 1$ T and it progressively saturates around 35% at 6 T. When the field is subsequently decreased, the loss does not come back to its initial value but it stays around 30% in zero field. Only further cycling to room temperature allows the recuperation of the initial loss. This microwave absorption below the transition could be due to a volume fraction of the sample which does not go superconducting. The influence of a magnetic field on this fraction will require additional measurements, especially frequency effects below the transition because relaxation phenomena are possible in the superconducting phase.

If the microwave loss may be interpreted adequately, the frequency shift is not easily understood. This frequency shift at the transition has been interpreted as an inductive effect on the microwave cavity for the case of granular aluminum films.[3] The extra inductance manifests itself by a frequency drop when the film becomes superconducting; this is caused by the inertia of the superconducting Cooper pairs working against the accelerating electric field. According to the first London equation,[10] a kinetic inductance appears, which is inversely proportional to the density of superconducting pairs. The frequency shift data of Fig. 1 could then potentially be used to deduce

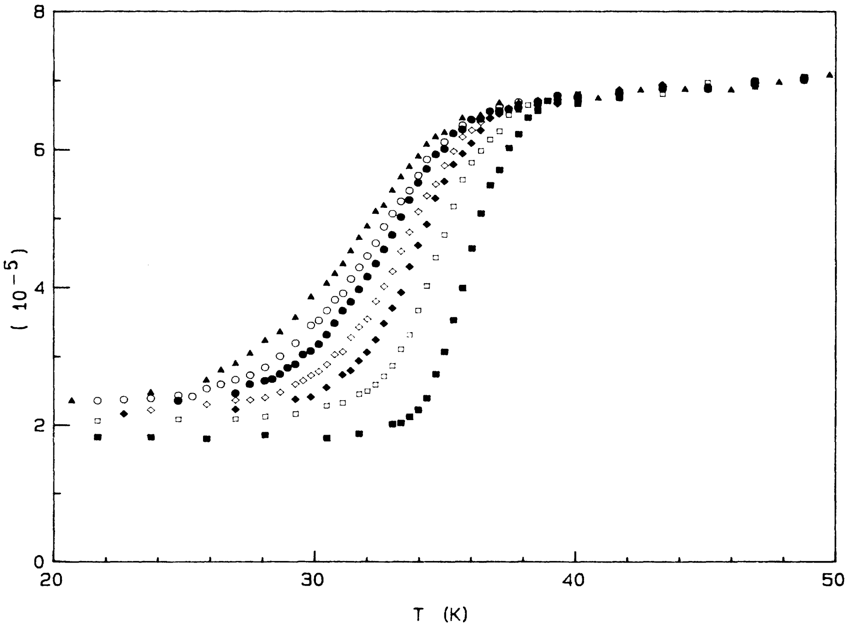

FIG. 3. Microwave loss $\Delta(1/2Q)$ at 16.8 GHz as a function of temperature: ■, 0; □, 1.07; ◆, 2.13; ◇, 3.17; ●, 4.24; ○, 5.31; ▲, 6.35 T.

such a density.

In summary, we have shown that the microwave measurements in a cavity are highly interesting to characterize the superconducting transition in high-T_c superconductors. Unlike dc transport measurements, the microwave data may give information about the superconducting state well below the transition temperature, but for the moment no precise model is available to analyze these data. The ac conductivity is determined with precision and magnetic-field effects on the transition are easily measured. The technique is particularly helpful to possibly determine the portion of the sample which is transforming and to study possible frequency effects in the superconducting state. These measurements are thus potentially very useful to characterize the nature of the superconducting state in these compounds.

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