EDDY CURRENT MEASUREMENTS OF THE NEW HIGH Tc CERAMIC SUPERCONDUCTORS

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INTRODUCTION

The discovery of ceramic superconductor materials which have transition temperatures above that of liquid nitrogen has inspired very active research in this area. These materials have great potential; however, there are many technique problems with fabricating them in useful forms. A major problem is that most fabrication schemes produce consolidated forms with relatively low critical currents. Successful fabrication of these materials requires a means of measuring the critical parameters in ways that can be used for process monitoring and control. In particular, the superconducting critical parameters (critical temperature Tc; current, Jc; and magnetic field, Hc) need to be measured in a nondestructive manner, preferably with a noncontacting, imaging technique. Accomplishing this goal is a new challenge to the NDE community. This paper is a beginning to the process of developing appropriate NDE diagnostic tools for these superconductors.

The material studied is YBa2Cu3O7-x prepared by two techniques: solid state reaction and sol-gel synthesis. These two techniques produced materials with approximately the same transition temperature but with significantly different homogeneities and eddy current responses.

EDDY CURRENT METHOD

The measurement consisted of inducing currents in the superconducting material and recording the induction they caused. Two nearly identical coils were used, as shown in Figure 1, in order to cancel the self inductances and temperature dependence of the coils.
The coils consist of 100 turns each of copper wire on nylon forms connected to a copper plate as shown. Since the skin depth for copper is about 0.0006 mm at 10 kHz, the copper plate effectively isolates the two coils electromagnetically. The sample, in the form of a pressed disc or pellet, is placed in one coil and in contact with the copper plate. The thermometer and heater were placed as in the figure to record the sample temperature and control the temperature drift rate. The assembly is covered with a copper cylinder and placed in a stainless steel outer can, which in turn is placed in a liquid nitrogen dewar. The purpose of the copper plate and cylinder is to ensure uniform temperature throughout the sample. With this arrangement a drift down in temperature to 80 K from 300 K takes around 30 minutes.

The coils form two arms of a standard Wheatstone bridge with the other two arms resistive. The bridge is excited in a continuous wave manner with excitation voltages of 0.3 to 3.0 Vrms, corresponding to magnetic field strengths at the sample center of around 0.001 to 0.1 Oersteds. These are very small values. Electrically, this method measures the difference in inductance of the two coils, which is balanced to zero by the bridge above the sample transition temperature, here at 100 K. When the sample goes superconducting, very strong diamagnetic currents are established in the material in order to shield the sample interior from the externally imposed magnetic field. This effect is related to the Meissner effect, which is fundamental to superconductivity, whereby the material acts to exclude all magnetic fields from its interior. The induced currents oppose the excitation currents by producing the opposite field within the coil. The impedance of the coil with the sample then is reduced when the sample goes superconducting. The balanced bridge is therefore offset by the introduction of an additional positive resistive and negative inductive impedance due to the supercurrents in the material. This offset voltage $V$ is measured with a lock-in amplifier synchronized to the excitation source.

The eddy current technique described is similar to the AC magnetic susceptibility measurement, which is normally done with separate excitation and pickup coils in long solenoidal design. This latter technique is quantitative in that the excitation field is uniform over the sample to a high degree and with small samples the sample shape is not important. The correspondence between the two techniques is

$$\text{Real}(V) \propto \omega \chi'$$  and  $$\text{Imag}(V) \propto \omega \chi''$$

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where $\chi', \chi''$ are the real and imaginary susceptibility components. Both techniques measure the average magnetization flux within the one coil brought about by the sample; the excitation flux is canceled by the empty coil. The eddy current technique described here lacks some precision in that the excitation field is not uniform in either direction of magnitude over the sample - some of the field lines cross the circumferential surface. However, the geometry used here is more easily adaptable to be used as an NDE tool by placing it above the sample or some other similar position, which would allow scanning in a noncontacting manner.

**SUPERCONDUCTING DIAMAGNETISM**

The basic response of a superconductor to an eddy current measurements can be illustrated with the use of London's equations [1]. Assuming the conduction properties of the material are isotropic, the supercurrent density is proportional to the magnetic vector potential, $A$, by

$$J = -(1/\mu_0 \lambda^2) A$$

where $\mu_0$ is the magnetic permeability, $B$ the magnetic induction, $E_{\text{vec}}$ the electrical field, and $\lambda$ the London penetration depth is given by

$$\lambda = \lambda_0 \left(1 - (T/T_c)^4\right)^{-1/2}.$$  

The temperature dependence for this penetration depth is empirically based on the previously known superconductors [2]. The value of the intrinsic London depth ($\lambda_0$) for these high $T_c$ materials is thought to be of the order of $\lambda_0 = 0.1 - 0.2 \mu m$ [3]. Quite generally, the square of the London depth is inversely proportional to the superconducting electron density in the material. The observed value is large compared to those of the more conventional superconductors and indicates that only a fraction of the conduction electrons in these materials participate in the superconducting state. It is now known that the conduction in these materials is along two dimensional planes and does not involve all the conduction electrons. In a powder sample the conduction must be an average over the orientations of the grains. The materials studied here are sintered from powder material. Therefore, in the sense of averaging over all grain orientations, the samples were isotropic and the net magnetization expected is less than that which would be observed in a single crystal with the excitation field perpendicular to the conduction planes.

Coupling London’s equation with Maxwell’s equations for electrodynamics allows the net flux within the coils to be calculated from an equation, which describes the Meissner effect:

$$\nabla^2 \mathbf{B} - (1/\lambda^2) \mathbf{B} = 0.$$

This equation is the same as that found for ordinary materials where the conduction current is $J = \sigma E$ and $\sigma$ is the electrical conductivity, if $1/\lambda^2$ is replaced by $i\omega \mu_0$ for continuous wave excitation. The simplest solution to the above equation that roughly corresponds to the experimental configuration is that of a cylindrical sample in an excitation field parallel to the symmetry axis. This solution yields a net magnetization for the two-coil setup which decreases very rapidly from 0 for $T = T_c$ to -1 (SI units) at $T = 0$. For reasonable values of $\lambda_0$, as quoted above, this drop occurs within a very narrow temperature region near $T_c$. However, this calculation assumes a continuous homogeneous superconductor, for which this measurement could be used to determine $\lambda_0$. In practice, it is very difficult to determine $\lambda_0$ this way due to its small value [2].
For these high $T_c$ materials it might be more appropriate to assume that they are made up of a large number of small particles (grains), each of which is superconducting [4]. The sintered pellet then contains individual isolated grains as well as grains interconnected by sintering and grain boundaries. In this model the excitation magnetic field would induce currents totally within, as well as between, grains or particles. Both currents would be measured by the eddy current technique. Only the intergrain current is important for many applications and it is this boundary that is currently thought to be the limitation to achieving high critical currents in consolidated bulk forms. The small particles or grains in the pellet would greatly affect the measured magnetic susceptibility. This is illustrated by calculating the magnetization expected for a pellet made up of unconnected spherical grains [1]. The net magnetization of the sample is proportional to the total sample mass (number of grains), the average orientation of the anisotropic grains, and a function dependent on the grains radius, $r$, and the London depth, $\lambda$, given by

$$\chi' = 3\coth(r/\lambda)/(r/\lambda) - 3/(r/\lambda)^2 - 1.$$  

This function is plotted in Figure 2 for three values of $r/\lambda$. It can be seen from the penetration of the excitation field to the depth $\lambda$, that the net magnetization is greatly reduced when the size of the grain is comparable to the London depth. Also, as grain size decreases, the temperature dependence of the magnetization becomes significantly less rapid. Since the London penetration depth is an intrinsic parameter of the superconducting material, the eddy current measurements directly reflect sample superconducting grain or particle size and to some extent intergrain connection.

RESULTS

Figure 3 shows the measurements for a sintered and oxygenated pellet made from the sol-gel derived powders. The sol-gel process produces powder particles with very controlled and homogeneous stoichiometry, hence single phase materials. The resistive transition of this pellet, measured by the AC four probe technique, is shown in Figure 3 to be quite narrow and smoothly varying. The figure also shows the real (in phase) and imaginary (out of phase) eddy current signals measured simultaneously with the resistive measurements. The large diamagnetism of the pellet is evident in the imaginary signal; the onset occurs at the same temperature at which the resistance begins to drop. The steep drop in the diamagnetic signal does not begin to occur until the resistance is near zero (experimentally). The figure also shows the real part of the eddy current signal, which corresponds to dissipation of the currents in the material. The reason for the peaked behavior of the real part of the eddy current signal is thought to be dissipation occurring when the applied field is equal to $H_{c1}$, the field at which flux lines begin to enter the material [5]. This field strength is reached for any excitation field as the transition temperature is reached, because $H_{c1}$ becomes zero at $T_c$. The temperature at which the peak dissipation occurs should then decrease as the excitation field strength is increased, which is observed even for the very small excitation field strengths used here.
Fig. 2. Calculated diamagnetism due to a spherical homogeneous isotropic superconducting particle with different values of the ratio of particle radius to intrinsic London penetration depth ($r/\lambda_o$).

Fig. 3. Eddy current response (imaginary and real parts) and resistance recorded for the YBa$_2$Cu$_3$O$_{7-x}$ pellet made by the sol-gel process. The measurements were made at 10 kHz and with a maximum field strength of about 0.12 Oersteds (rms).
Figure 4 shows the experimental results for a sample pellet prepared by the solid state reaction technique of mixing powders and sintering. This sample is known to be less homogeneous than the sol-gel sample, which is brought out both in the resistance and eddy current signals. The resistance drop shows a distinct change in slope and the resistance reaches zero at a considerably lower temperature. This behavior could indicate a second phase in the material that superconducts at a slightly lower temperature [5]. The eddy current signals are also markedly different from those of the sol-gel sample. In particular, the diamagnetic signal decreases much slower with temperature, which could be indicative of a smaller effective value in the superconducting particle or grain size. Finally, the dissipation also differs from that found for the sol-gel sample in that it shows two peaked and much broader regions. All these results indicate that the material prepared by the solid state reaction is considerably more inhomogeneous than that prepared by the sol-gel technique.

The microstructures of these materials were found to be extremely complex, see figure 5. The grains were platelet in form, due to the anisotropic crystal structure of this material. There was random orientation of the grains and porosity both between the grains and in relatively large isolated regions. The nature of the interface between the grains is not known. Optical micrographs of the two samples showed the sol-gel material to possess grain platelets typically 3 μm thick and 10-20 μm long and wide. The grains for the solid state reaction material were similar in shape but much larger in size with thicknesses of around 10 μm and lengths and widths of 30-50 μm. On the basis of the spherical grain model presented, the eddy current response of the two materials appears to be switched around. The reduced diamagnetism and temperature dependence of the imaginary response for the solid state reaction material would indicate that it has smaller superconducting particles than the sol-gel material. Either the model is too simplified to be valid for these materials or only a small part of each grain is participating with the supercurrent for the solid state reaction material. Some preliminary experiments with other solid state reaction samples show a drastic drop in the diamagnetism when they come in contact with water. Water is known to alter the oxygen content of these materials and produce a nonsuperconducting phase. This conversion would decrease the size of the superconducting portion of a grain with out changing the size or shape of the grain as a whole. Other phases of the material, which would not show diamagnetism, are also likely present in these materials.

CONCLUSIONS

The difference between magnetic susceptibility and eddy current measurements is mainly that of geometry and applied field uniformity. The ability of the eddy current technique to be performed in unusual geometries makes it a versatile and useful tool for characterizing superconducting materials. This technique is noncontacting and can be made to provide images of responses over the surface of materials that should be very useful for future process control. The calibration of the eddy current response must be related to the standard AC magnetic susceptibility technique, but this should not present major problems if suitable geometries are found which maximize the uniformity of the applied field for a given situation. The response of two high Tc samples prepared by different techniques has been presented. The responses were found to be very dependent on the material microstructure and processing technique. A detailed explanation of the eddy current response must further address the effects of microstructural parameters
Fig. 5. Photographs of the microstructure for the sol-gel prepared (a) sample and the sample prepared by solid state reaction (b). Magnification is 600X.
Fig. 4. Eddy current response (imaginary and real parts) and resistance recorded for the YBa$_2$Cu$_3$O$_{7-x}$ pellet made by the solid-state reaction process. The measurements were made at 10 kHz and with a maximum field strength of about 0.12 Oersteds (rms).

such as grain size, porosity, phase composition, and particle interconnection. The effects of many of these parameters should be measurable in a rapid manner by the eddy current technique.

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REFERENCES


