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Millimeter wave Absorption of Y—Ba—Cu Based High Tc Superconductors

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ABSTRACT

Microwave absorption of the Yttrium-barium-copper oxides have been measured by using an open resonator technique in the temperature range of 20 K to samples' critical temperatures and beyond. The samples with the start stoichiometries characterized as $Y_4Ba_7Cu_{11}O_x$ $Y_5Ba_6Cu_{11}O_x$ $YBa_2Cu_3O_x$ and $Y_2Ba_4Cu_8O_x$ have indicated a sharp drop in their temperature dependent surface resistance or the normal to superconducting phase transition with the range of 84 to 92 K, and the "123" sample shows a highest Tc as well as a lowest residual Rs, which is shown not only an intrinsic property of those materials but is associated with impurity phases or interfaces. Significant absorption in the understudied materials was observed at temperature well below Tc, which may be associated with the intrinsic behavior of the materials despite of its sample dependent. We suggest that the anisotropy showing both in structure and electromagnetic behavior of these materials might be one of the dominate factors.

INTRODUCTION

Measuring microwave absorption is a very sensitive technique for studying phase transitions which involve sharp change in the conductivity or the dielectric constant. Several studies of high Tc superconductors have focused on measuring the temperature dependence of the absorption of electromagnetic waves [1-5]. For YBCO ceramics, the results of these measurements typically show a microwave absorption that drops rapidly below the normal state as the temperature decrease below Tc, but at low temperatures there remains a significant residual loss which seems to be somewhat sample dependent. On the other hand, using two powdered samples with grain sizes< 5um. some workers have found that the absorption became negligible within a few degrees of Tc [6]. In order to understand the reason for this discrepancy, it was decided to make a systematic study of the dependence of microwave absorption on different stoichiometric proportions of the metallic ions .

We have made microwave absorption measurements on large number of yttrium-barium-copper based oxides with the start stoichiometry as :$YBa_2Cu_3O_x$, $Y_2Ba_4Cu_8O_x$, $Y_5Ba_6Cu_{11}O_x$, $Y_4Ba_7Cu_{11}O_x$. The samples used in the present study were prepared by the conventional ceramic heat-grind-heat etc. technique. The transition temperature occurred within 84K t0 90K for all of our samples, as described by the onset of Meissner effect measured by the ac susceptibility shown in Fig.1 .

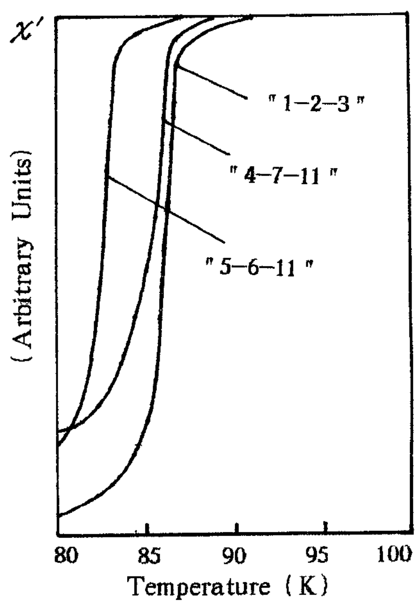

FIG.1 ac susceptibility of the Y—Ba—Cu based oxides.

SURFACE RESISTANCE CHARACTERIZED METHOD

A complete description of the microwave measurements technique has been described elsewhere [3]. In brief, the samples were mounted

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as one end plate of a quasi-optic coupling open cavity operating in the 'TEM_{00q} mode (Being an integer, q denotes the number of wave-nodes along the cavity's symmetric axis) resonating at 95 GHz. At the temperature T_i, we measure the frequency dependence of mode's reflecting power Pr and adapt a nonlinear least-square technique to fit the expression (1a). As a result the coupling cofficience β , loaded quality factor Q_l and resonant frequency f_0 can be fitted out.

$$P_r(\Delta f) = P_i \left[1 - \frac{4\beta}{(1+\beta)^2} \frac{1}{1 + 2(Q_l \Delta f / f_0)^2}\right] \quad (1a)$$

Where P_i and f_i are input power and frequency respectively, and f is given by $\Delta f = f_i - f_0$. Consequently, the change in the mode's bandwidth and frequency as a function of temperature with the sample mounted compared with a polished copper endplate could be decided. The difference of Rs and Rcu is proportional to that of resonant bandwidth, and it is given by:

$$R_s - R_{Cu} = G\,(\Delta\omega_S - \Delta\omega_{Cu})/\omega_{Cu} \quad (1b)$$

where G denotes the geometric factor determined from the resonant mode and resonant cavity's characteristic dimensions.

RESULTS AND DISCUSSION

First, consider the "1-2-3" composition sample; As already shown, the onset of superconductivity is marked by a sharp drop in Rs shown in Fig.2a. The transition are quite sharp, having width of the order of 6 K which are comparable to those observed in DC resistivity. This sample shows a lowest residual resistance at low reduced temperature compared to the others .

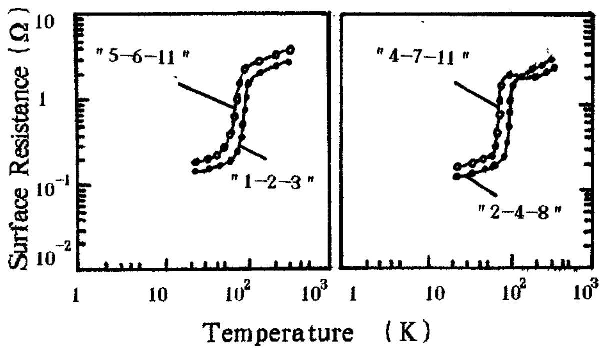

FIG. 3 Temperature dependence of surface resistance for Y-Ba-Cu superconducting ceramics.

Fig.2b exhibits the behavior of "4-7-11" sample, which associated with the semiconducting nature of Y_2BaCuO_5 , but is strangely followed by a sharp drop at temperature near 86 K, which seemed to be attributed to the change in the Cu average valence. This sample also shows a higher Rs level than that of "123" sample. As we have seen, magnetic measurements indicates that this sample contains few percent of high-Tc phases such as 123 phase; Hence,we might suggest that spurious phases such as barium cuprate and 211 phase should greatly affect Tc and Rs. Besides,the result is a good demonstration of the capability of our technique to detect small fractions of superconducting inclusions in a predominantly insulating material.

Again from Fig.3, all the samples measured, whether its start composition is characterized in 1-2-3 or not, have shown a normal to superconducting transition near within 84 92 K. The result as well as our R-T curve and susceptibility measuring tells us that the superconductive transition at 90 K in these Y-Ba-Cu based compounds may have little dependence on the material's start stoichiometry, but mainly and may only on the structure of crystal cell. A similar behavior for "248" sample to that for "123" one could be seen from this figure. But the late exhibits a more favor characteristic in higher Tc and less microwave absorption, and we have reasons to believe that the "123" phase in the sample is most favor to the high Tc superconducting phase than the others.

The surface resistance of these superconductors at 95 GHz and at low reduced temperature,say t = T/Tc = 0.3 , is only of the same order (slight higher) of magnitude as that of copper under the same conditions, which is inconsistent with the prediction of BCS theory. This over large value relative to the conventional superconductors may arise from several factors. First, portions of sample such as the spurious phases may be normal independent of temperature. Second, grain boundaries may also impede the microwave current and further raise the surface resistance. Also, the strong pinning effect may lead the ceramic to exist preferly in the surface state instead of Meissner state. As a result the microwave current is forced to flow along the separated passages which are the so-called superconducting channels. If any such ways cross the lattices of the material,then the weak link of Josephson junctions in the material would be partly interrupted or destroyed, which might introduce some additional absorption [7-9].

Finally, in this Rs-T figure, nearly the same slope of each curve shows that there may be some intrinsic mechanisms governing the electromagnetic behavior of Y-Ba-Cu based superconductor, and we think of the anisotropic behavior as one of the dominate factors [10].

For simplicity, we first consider the interaction of a plane wave with a half-infinitive isotropic superconductor plate located at XOY plane shown in Fig.3

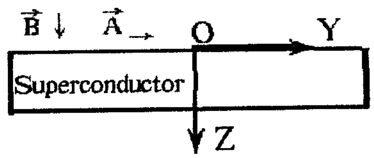

FIG. 3 Co-ordinates chosen for further analyzing the behavior of superconductors in microwave field.

The wave inciding on the interface will be partly reflected and partly absorbed. The surface resistance of the superconducting plate can be defined as $Z^{(s)} = (Et/Ht)|_{z=0}$, where subscript "t" means the transverse. We assume that this plane wave vary with time t and z axis in the form of $\exp(i\omega t - ikz)$. We know that waves in superconductors should be governed by Maxwell Eqs. and material's constitutive Eqs. arised from London or Ginzberg-Landau theory. Made some essential mathematic procedures, we can get the impedance'expression by :

$$Z^{(s)} = -\omega\mu_0/k \qquad (2)$$

where μ_0 is the permeability of the free space, $\delta_n = 1/(\omega\mu_0\sigma_n)^{1/2}$ is the skin depth and k is the wave number given by :

$$k^2 = -(1 + i\lambda_L^2/\delta_n^2)/\lambda_L^2 \qquad (3)$$

For $Re(Z^{(s)}) > 0$, the positive sign was chosen. In superconducting state, the inequality $\lambda_L << \delta_n$ holds, and taking first order approximation,we come to the well known expression for $Z^{(s)}$:

$$Zs = \omega^2\mu_0^2\sigma_n\lambda_L^3 + i\omega\mu_0\lambda_L \qquad (4)$$

But for superconducting oxides,the situation is quite different. Its strong anisotropy showing both in the structure and physical features makes the interaction of electromagnetic waves with superconducting electrons having some unique feature to be characterised. It is known that for $YBa_2Cu_3O_x$ material, the orthehombic phase shows a superconducting phase transition at the temperature near around 90 K . We may chose the coordinate axes $\vec{a}_x, \vec{a}_y, \vec{a}_z$ coincide with the lattices axes a , b , c on supposing that crystal lattices are highly oriented). Assume that the effect relative dielectric constant along $\vec{a}_x$, $\vec{a}_y$, $\vec{a}_z$ are $\varepsilon_{rx}, \varepsilon_{ry}, \varepsilon_{rz}$ respectively.

Now let us deal with the effect of anisotropy on a plane wave propagating along the direction of the unit vector $\vec{n} = n_x\vec{a}_x + n_y\vec{a}_y + n_z\vec{a}_z$ in the form of $\exp(-i\omega t - ik\vec{n}\cdot\vec{r})$ where the wave number is to be found out. From Maxwell's Eqns. , get:

$$-i\omega\mu_0\vec{H} = \nabla \times \vec{E}_0\exp(-ik\vec{n}\cdot\vec{r}) = -ik\vec{n} \times \vec{E} \qquad (5a)$$

$$i\omega\vec{D} = (ik/\omega\mu_0)[\vec{E} - \vec{n}(\vec{n}\cdot\vec{E})] \qquad (5b)$$

Expressing $D_j/\varepsilon_0\varepsilon_{rj}$, for j=x,y,z, we have:

$$\vec{D} = \frac{k^2}{k_0^2}\varepsilon_0\vec{n}\cdot\vec{E}\left(\frac{n_xk_x^2}{k^2 - k_x^2}\vec{a}_x + \frac{n_yk_y^2}{k^2 - k_y^2}\vec{a}_y + \frac{n_zk_z^2}{k^2 - k_z^2}\vec{a}_z\right) \qquad (5c)$$

From Eqn.(5b), we know $\vec{n}\cdot\vec{D} = 0$, hence,

$$\frac{n_x^2\varepsilon_{rx}}{k^2 - k_x^2} + \frac{n_y^2\varepsilon_{ry}}{k^2 - k_y^2} + \frac{n_z^2\varepsilon_{rz}}{k^2 - k_z^2} = 0 \qquad (5d)$$

with $\varepsilon_{rj} = k_j^2/k_0^2$ for j = x, y, z and $k_0 = \omega^2\mu_0\varepsilon_0$.

It is eigen-equation for wave number k in an anisotropic material from which the two roots of are determined. It is confirmed that ε_{rj} (j=x,y,z) is a function of the London penetration depth . For simplify we may chose $|\varepsilon_{rx}| \approx |\varepsilon_{ry}| >> |\varepsilon_{rz}|$; Hence,it is necessary for k to obey the below inequality:

$$k_0\varepsilon_{rx} >> k_0 > k_0\varepsilon_{rz} \qquad (5e)$$

Some interesting results may be drawn out from Eqn.(5d) and Eqn.(2). Despite of its quasi-two dimensional behaviors, in YBCO the interaction between ledges should play some important role, which is confirmed through many experiments. Here, it can be further indicated by examining the the fact that direction of the Pointig vector $\vec{E} \times \vec{H}^*$ is no longer along that of $\vec{n}$. This means some interactions between the longitude and transverse components of wave function. It is believed that the poor conducting behavior along C axis, which corresponds to a small wave number value, is doomed to affecting the behavior of a-b plane [10]. In detail, the solution for wave number of Eqn.(5) must far less than $k_0\sqrt{\varepsilon_{rx}}$ the value corresponding no anisotropy being taken into consideration. Therefore, the surface resistance level of this plane is forced to be raised.

It is no double that the Rs of highly oriented YBCO thin films should be several orders lower than that of ceramics because its less impurity, high orientation which is helpful for us to get rid of the effect of the poor conducing behavior along c-axis. However, because of the difficulties in preparing the highly oriented bulk samples using a conventional ceramic heat- grind-heat technique it is hardly possible for us to make Rs of such bulk materials to be as small as that of the C-oriented YBCO thin films. On the other hand, the local disorder in Cu-O chains as well as the existence of the interaction between the longitude and transverse components of wave function mentioned above are obstacles for us to obtain the high-Tc materials with the magnitude of Rs several orders (say 4 or 5) lower than that of OFHC copper.

Again, at low reduced temperature the measured surface resistance does not saturate to a low, constant value as would be expected from an isotropic BCS superconductor with a thermally activated temperature dependence of σ ,which might partly due to the nonlinear response of material to electromagnetic waves.

The nonlinear response must be taken into account , because of the strong anisotropy of the material, which could cause the change of eigenstate in the material, as shown in Eq.(4) that a change in wave number k. With the ansatz $\vec{H}(\vec{r},t)=\vec{H}(\vec{r})f(t)$, Eqn.(2) results in:

$$Z^{(s)}(\vec{r}\ ,\ t)f(t)=L_s\frac{\partial f}{\partial t}+R_s\cdot f(t) \qquad (6)$$

A vector $|\vec{H}(\vec{r})$ dependence of $Z^{(s)}$ can only be a dependence on the combination $\vec{H}(r)\cdot\vec{H}^*(r)=|\vec{H}(r)|^2$, as long as there exist no preferential direction in the material . Therefore, at low field i.e. $H \ll \min(H_c^c\ ,\ H_c^{a-b})$, we can develop $Z=Z(\vec{H}^2(r))$ in a power series in $h^2(\vec{r})=\vec{H}\ \vec{H}^*/H_c^2$ with $H_c=\min(H_c^c\ ,\ H_c^{a-b})$,and Rs is given by:

$$R_s(r,t)=R_0\ [1+\alpha\ h^2(\vec{r})f^2(t)+\frac{1}{2}\gamma\ h^2(\vec{r})] \qquad (7)$$

where α describes effects fast compared to an rf period, whereas γ
includes various slow effects; and the higher orders are omitted. The estimate of $\alpha(\omega)$ is complex and difficult. The main parts in α are the result of the additional absorption due to the "lowered" energy gap and due to the possibility of multiphonon absorption. The world "lowered" is somewhat the mean that some energy states might exist in the energy gap, which is preliminary shown in temperature dependence of the surface resistance, and this intrinsic mechanism should also produce $\lambda_L(t)$ with a non-BCS form.

CONCLUSION

Using an open resonator with quasi-optic coupling, we have measured the temperature dependence of surface resistance of Yttrium-Barium-Copper oxides. The superconductive transition near 90K has been observed in samples whose chemical compositions are departure from that of $YBa_2Cu_3O_x$,which indicates that the start stoichiometry of the material is not so important to this high Tc phase as thought before. However, the spurious phases such barium cuprate and 211 phase have shown great effect on the microwave properties of the samples, and even for superconducting 123 and 124 phases the distinguishable Tc and Rs values have been observed. Our results indicate that the 123 superconducting phase is most favorable to high Tc and low absorption compared to the others. At very low reduced temperature over large residual resistance and the non-BCS form of Rs(T) were observed in all the measured samples, which may be associated with some intrinsic behaviors of the materials despite of its sample or impurity phases dependent. We suggest that the nonlinear effect caused by anisotropy might be one of the dominate factors.

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