## Penetration depth measurement in high quality $YBa_2Cu_3O_{7-x}$ thin films

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**Abstract.** The parallel plate resonator method has been used for measuring high quality  $YBa_2Cu_3O_{7-x}$  (YBCO) thin films, which have low temperature residual losses comparable to those previously obtained in single crystals. The surface resistance and the real part of the conductivity show a non-monotonic behaviour with a broad peak around 45 K. The penetration depth and the real part of the conductivity vary linearly at low temperatures. The lowest penetration depth linear fitting has a slope value of 2.2 Å/K to 2.5 Å/K up to 20 K which is lower than previous measurements on YBCO single crystals. An interpretation of this smaller slope in terms of the generally accepted *d*-wave order parameter symmetry presents difficulties.

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Determining the symmetry of the superconducting order parameter is one of the main issues in understanding the mechanism that causes superconductivity in the high  $T_c$  cuprates.

For a *d*-wave superconductor the energy gap vanishes along four lines in the  $k_z$  direction located at the position  $|k_x| = |k_y|$  on a cylindrical Fermi surface. These node lines lead to a change in the penetration depth as a function of temperature that varies linearly with temperature, *i.e.*  $\lambda(T) \propto T$  [1,2]. In contrast, for an s-wave superconductor one has an exponential monotonic behaviour of the superconducting properties [3,4]. In the case of YBCO single crystals, the linear variation of the penetration depth at low temperatures is accompanied by a non-monotonic variation of the surface resistance  $R_s(T)$  and of the real part of the conductivity  $\sigma_1(T)$ . By introducing Zn impurities into a  $YBa_2Cu_3O_{7-x}$  (YBCO) single crystal, Bonnet et al. [6] were able to show the suppression of that nonmonotonic behavior and did get a change of  $\Delta\lambda(T)$  from a linear to a quadratic temperature dependence, as predicted by d-wave theory. More generally, the microwave surface impedance  $Z_s = R_s + i\omega\mu_0\lambda$  yields important information regarding the superconducting carrier density  $n_s(T)$  and the quasiparticles scattering rate  $1/\tau(T)$ , whose low temperature limit can give us precious indication about the quality of the high  $T_c$  material. In low quality films,  $\tau(T)$  reaches very fast its maximum value close to  $T_c$  [7].

There are two contributions to losses in YBCO thin films at microwave frequencies regime, one which has an intrinsic origin and the other one an extrinsic one. One of the long standing problems is the residual surface resistance [5] which has presumably an extrinsic origin such as twins, grain boundaries and other defects. These extrinsic losses can smear out the non-monotonic behaviour of  $R_s(T)$  in YBCO single crystals [6], and they can influence in the same manner YBCO thin film properties. It was shown that this non-monotonic behavior manifests itself as a broad peak in  $R_s(T)$  around 40 K which is more conspicuous in  $\sigma_1(T)$  [7]. This broad peak can be an indication for a low defect density sample, where the scattering time  $\tau(T)$  attains a higher value at low temperatures. It can be understood in the framework of the two fluid model by using the Drude formula  $\sigma_1 \propto n_{qp}\tau$  in the limit  $\omega\tau \ll 1$ , where  $n_{qp}$  is the quasiparticles density.

A non monotonic behaviour of  $\sigma_1(T)$  was reported by Ma *et al.* [8] in YBCO thin films, but it was accompanied by  $\Delta\lambda(T) \propto T^2$  in the low temperature range. On the other hand, Hardy *et al.* reported a linear behaviour  $\Delta\lambda(T)$  of for a high quality YBCO single crystal [2], showing the non-monotonic behaviour of  $\sigma_1(T)$ .

In our experiment we measured high quality YBCO thin films on LaAlO<sub>3</sub> substrates. These films where grown by pulsed laser deposition at 785 °C and 300 mtorr of molecular oxygen [9]. The thickness of these films is about 3000 Å, their  $T_c$  is ~ 91.5 K and a transition width of 0.3 K is obtained by four point resistive measurement. The best pair of such films used in our parallel plate

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resonator gave a small residual loss of about 50  $\mu\Omega$  at 10 GHz. These residual losses are intermediate between two values reported for YBCO single crystals (200  $\mu\Omega$  [10],  $10 \ \mu\Omega$  [11] at 10 GHz). They are approximately lower by a factor 2 than the residual losses reported by Ma et al. [8] for their lower  $R_{res}$  post annealed YBCO thin films. The parallel plate resonator technique [12] (PPR) has two major advantages in comparison to other resonant cavities. The surface resistance can be calculated directly from the measured Q-value without any corrections. It is a highly sensitive method for measuring small changes in the penetration depth, although it does not lend its absolute value. In our measurements we used two  $1 \times 1 \text{ cm}^2$  YBCO thin films with a Teflon spacer 25  $\mu$ m thick in between them. We rely on a weak coupling transmission mode signal. The PPR was measured using the HP-8510C vector network analyzer.

The change of the penetration depth and of the real part of the conductivity can be calculated from the phase velocity of the electromagnetic field in the PPR and is given by [13,14]

$$V_{\Phi} = \frac{1}{\sqrt{\varepsilon\mu_0}} \frac{1}{\sqrt{1 + \frac{2\lambda}{s} \coth(\frac{t}{\lambda})}} \tag{1}$$

where  $\lambda$  is the penetration depth, t the film thickness and s the dielectric spacer thickness. In the low temperature limit, where  $\lambda < t$ , one can get the change of the penetration depth directly from the measured frequency shift of the PPR,

$$\Delta \lambda = \frac{s \left[ \frac{f^2(T_0)}{f^2(T)} - 1 \right]}{2 \left[ \coth\left(\frac{t}{\lambda(0)}\right) + \frac{t}{\lambda(0)} \sinh^{-2}\left(\frac{t}{\lambda(0)}\right) \right]}$$
(2)

where  $T_0$  is the temperature value from which the change of the penetration depth is measured (here about 5 K). Once the surface resistance and the penetration depth are known one can get the real part of the conductivity from the following equation [13,14]

$$\sigma_1 = \frac{2R_s}{\lambda^3 \left(\omega\mu_0\right)^2} \left[ \frac{\sqrt{1 + \frac{2\lambda}{s} \coth\left(\frac{t}{\lambda}\right)}}{\coth\left(\frac{t}{\lambda}\right) + \frac{t}{\lambda} \sinh^{-2}\left(\frac{t}{\lambda}\right)} \right].$$
 (3)

The surface resistance calculated directly from the measured Q is shown in Figure 1. The lower curve was obtained for a couple of high quality YBCO thin films with the lowest residual losses. The non-monotonic behaviour, expected for high quality YBCO, is not so prominent in the  $R_s(T)$  measurement, but appears clearly in  $\sigma_1(T)$  as shown later. The upper curve was obtained for lower quality YBCO thin films. It shows a more monotonic  $R_s(T)$ dependence. The residual losses are displayed more clearly in Figure 2. They are quite low for the lower curve (sample A),  $R_{res} \leq 50 \ \mu\Omega$ . For the upper curve (sample B)



Fig. 1. The surface resistance at 10 GHz of two YBCO thin films pairs grown on LaAlO<sub>3</sub> by PLD technique.



Fig. 2. The two YBCO pair films residual surface resistance measured at 10 GHz.

 $R_{res} \leq 150~\mu\Omega$ . The origin of the  $R_{res}$  in YBCO is not well understood. In one case a small reduction of  $R_{res}$  in YBCO single crystal is reported when introducing Ni or Zn impurities [7]. In another case it was shown that irradiation of two YBCO thin films by 25 MeV O<sup>16</sup> ions [15], suppresses the  $R_s$  non-monotonic behaviour. One of the films showed a decrease in  $R_{res}$  after the first irradiation, while in the second one  $R_{res}$  increased. Further irradiation leads  $R_{res}$  to increase in both films. In another work it was shown that the  $R_{res}$  can increase due to lack of oxygen [16].

Table 1 shows  $R_{res}$  values together with the change of the penetration  $\Delta\lambda(T)$  in the low temperature range for several YBCO samples. The third column represents the total change of the penetration depth  $\Delta\lambda = \lambda(T) - \lambda(T_0)$ . For sample A we have assumed  $\lambda(0) = 1500$  Å, and for sample B,  $\lambda(0) = 2000$  Å. As one can see,  $\Delta\lambda$ tends to increase with increasing  $R_{res}$ . Figures 3a (sample A) and 3b (sample B) show the temperature dependence of the penetration depth at the low temperature range.

Material	$egin{array}{l} R_{res} \ (10  { m GHz}) \end{array}$	$\lambda(T) - \lambda(T_0)  [{ m \AA}]$	The slope of $\Delta\lambda/T$ for the linear fit $[{ m \AA/K}]$	$\begin{array}{c} \mathbf{Growing} \\ \mathbf{method} \end{array}$	Reference number
YBCO/LaAlO <sub>3</sub> films (sample B)	$R_{res} \le 150 \ \mu\Omega$	$\lambda(20) - \lambda(5) = 54$	3.8	PLD	current work
YBCO/LaAlO <sub>3</sub> films (sample A)	$R_{res} \le 50 \ \mu\Omega$	$\lambda(20) - \lambda(5) = 32$	2.2	PLD	current work
YBCO/LaAlO <sub>3</sub> films	$R_{res} \le 800 \ \mu\Omega$	$\lambda(23) - \lambda(9.2) = 61$	4.42	dc sputtering	[17]
<i>in situ</i> YBCO films	$R_{res} \le 40 \ \mu\Omega$	$\lambda(20) - \lambda(5) \simeq 40$	$T^2$ dependence	-	[8]
<i>in situ</i> YBCO films	$R_{res} \leq 20 \ \mu \Omega$	$\lambda(20) - \lambda(5) = 47$	$T^2$ dependence	-	[8]
$ m YBCO/LaAlO_3$ films	$R_{res} \leq 140 \ \mu\Omega$	$\lambda(20) - \lambda(5) = 120$	$T^2$ dependence	BaF <sub>2</sub> post- annealed films	[8]
YBCO crystal	$R_{res} \leq 50 \ \mu\Omega$	$\lambda(20) - \lambda(5) = 68$	nearly linear-average slope $\simeq 4.5$	flux-growth technique	[18]
$ m YBCO/LaAlO_3$ thin film	$R_{res} \le 100 \ \mu\Omega$	$\lambda(20) - \lambda(8) \simeq 60$	exponential behaviour	dc sputtering	[20]
thin film	$R_{res} \leq 400 \ \mu\Omega$	$\lambda(20) - \lambda(6) \simeq 112$	exponential behaviour	dc sputtering	[20]

**Table 1.** The change of the penetration depth for various qualities of YBCO superconductors in comparison to the penetration depth change at low temperatures.



Fig. 3. (a) The temperature dependence of the penetration depth for sample A at the low temperature range. The slope of the linear fit is 2.2 Å/K. (b) The temperature dependence of the penetration depth for sample B at the low temperature range. The slope of the linear fit is 3.8 Å/K.

The two sets of data are related to the two  $R_s(T)$  curves presented in Figure 1. The slopes obtained from a fit to a linear temperature dependence are respectively equal to 2.2 Å/K for the lower  $R_{res}$  sample A, and 3.8 Å/K for the higher  $R_{res}$  sample B, in the temperature range from 5 K to 20 K. Actually, a better linear fit is obtained for sample A in the range from 10 K to 25 K, the slope is then 2.5 Å/K. For sample B, the data shows a positive curvature in the entire temperature range.

A linear penetration depth temperature dependence is predicted for a *d*-wave superconductor with lines of nodes [1],  $\Delta\lambda(T)/\lambda(0) \approx \ln(2) T/\Delta_0$  where  $\Delta_0$  is the gap value. This is in agreement with the slope reported for a single crystal, taking  $\Delta_0 = 22 \text{ meV} [2]$ . Impurities can alter this linear dependence to a quadratic temperature dependence one [7]. Unidentified imperfections can apparently cause the same quadratic behavior in YBCO thin films [1,8]. For an overdoped, high  $R_{res}$  YBCO thin film measured in our set-up a slope of  $\sim 4.3 \text{ Å/K}$  was obtained as shown in the fourth row of Table 1 [17]. The small slope reported here for the better sample is difficult to understand, if the linear behavior is interpreted as being due to a *d*-wave symmetry. As disorder is increased, the change of  $\lambda$  with temperature should become smaller,



Fig. 4. The real part of the conductivity measured at 10 GHz for sample A (open circles). One can see a broad peak around 45 K which is an indication for high quality of the thin films. This peak is absent for sample B (open triangles).

while it appears experimentally to become larger. A different interpretation of the linear behavior was proposed some time ago by several authors [21], as arising from phase fluctuations of the order parameter, but the calculated slope was smaller that the one measured on single crystals, by at least one order of magnitude; this interpretation was therefore discarded. Yet, the predicted slope varies as  $\lambda(0)^3$ , and is therefore quite sensitive to a weak disorder, surely present in the films, known to increase the value of  $\lambda(0)$ . An increase by 30% would be sufficient to obtain a slope of the order of 1 to 2 Å/K. Other possible explanations for the smaller slope, in the framework of a *d*-wave interpretation, are a larger gap [22,23], or the presence of a twin-related large *s*-wave component.

The non-monotonic behaviour of  $\sigma_1(T)$  is shown in Figure 4 for the best set of samples (calculated for  $\lambda(0) =$ 1500 Å).  $\sigma_1(T)$  has its maximum value at ~ 45 K in agreement with the results for a pure YBCO single crystal at 34.8 GHz [7]. This is also an indication for high sample quality. Yet, the smaller peak amplitude indicates a smaller value of  $\tau_{res}$ , as compared to the pure single crystal case [6]. We note (Fig. 4) the linear slope of the real part of the conductivity at low temperatures (below 20 K). This linear slope was measured also on an YBCO single crystal [18]. According to the theoretical prediction for a *d*-wave gap,  $\sigma_1(T)$  should vary as  $T^2$  at the microwave frequency range [19]. We show also Figure  $4 \sigma_1(T)$  for sample B, calculated for  $\lambda(0) = 2000$  Å.

In conclusion we have presented surface impedance measurements on high quality YBCO thin films. These films have residual losses comparable to those of single crystals. The linear  $\Delta\lambda(T)$  behaviour that we observe is in qualitative agreement with the *d*-wave superconducting model, but the slope that we observed in our best samples (2.2 Å/K to 2.5 Å/K) is smaller than reported for single crystals. A non-monotonic behaviour in  $\sigma_1(T)$  was observed for the best set of samples. It can be a good indication for the quality of the thin films. The linear  $\sigma_1(T)$  in the low temperature range is inconsistent with the quadratic temperature dependence predicted by the *d*-wave theory.

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