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Linear temperature variation of the penetration depth in $YBa_2Cu_3O_{7-\delta}$ thin films

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Abstract. – We have measured the penetration depth $\lambda(T)$ on $YBa_2Cu_3O_7$ thin films from transmission at 120, 330 and 510 GHz, between 5 and 50 K. Our data yield simultaneously the absolute value and the temperature dependence of $\lambda(T)$. In high-quality films $\lambda(T)$ exhibits the same linear temperature dependence as single crystals, showing its intrinsic nature, and $\lambda(0) = 1570$ Å. In a lower quality one, the more usual T^2-dependence is found, and $\lambda(0) = 3400$ Å. This suggests that the T^2 variations is of extrinsic origin. Our results put the d-wave–like interpretation in a much better position.

The puzzling properties of high-T_c superconductors have stimulated recently extensive experimental and theoretical work in order to determine the actual symmetry of the order parameter. While in the BCS weak-coupling theory, the penetration depth $\lambda(T)$ varies as $\exp[-\Delta/k_BT]$, a quadratic behaviour $\lambda(T) \sim T^2$ was reported by many groups in YBCO thin films [1]-[3]. Recently, in very high-quality YBCO single crystals, a linear temperature dependence was measured up to 40 K [4]-[6]. Such a variation, never observed so far in YBCO thin films[1], is consistent with the occurrence of nodes in the gap and may suggest a d-wave pairing mechanism [8]. However, other possible interpretations have been raised, both theoretically and experimentally, *e.g.* a possible proximity effect with a normal metal layer [6], [9], or the sensitivity of a conventional BCS-type $\lambda(T)$ to the oxygenation of the samples [10], [11]. Several reasons for the discrepancy observed between thin films and single crystals

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[1] Beasley [7] reports a linear temperature dependence on one TlBa-CaCuO thin films.

may be invoked: i) within the framework of d-wave pairing, scattering due to impurities or defects may change the linear temperature dependence into a quadratic one in thin films [12], where such defects would be more numerous. However, T_c should then also be affected, which is not the case [4], [13]. ii) Weak links, more likely to be present in thin films, may yield an effective penetration depth (larger than the intrinsic one) with a different temperature dependence [14], [15]. The penetration depth λ has been essentially measured in both single crystals and thin films by using surface impedance techniques at a single frequency [2], [4]. Such measurements actually measure the variation $\Delta\lambda(T) = \lambda(T) - \lambda(0)$, but without the knowledge of $\lambda(0)$ on each sample, comparing the temperature dependence of various samples may not be significant. This paper reports for the first time, from high-frequency transmission data, both an unambiguous linear temperature dependence of $\lambda(T)$ on high-quality YBCO thin films, which is quantitatively the same as in single crystals [4], and simultaneously $\lambda(0) \sim 1570$ Å. We observe a quadratic temperature dependence on a lower-quality sample, and $\lambda(0) \sim 3400$ Å. We suggest that this T^2-dependence in thin films is not a consequence of strong scattering (in contrast with Zn- or Ni-doped materials [6]) but is of extrinsic origin. The intrinsic dependence of $\lambda(T)$ at low temperature is linear. This linear dependence points plainly toward the existence of low-energy excitations, but our results alone do not allow to identify them. A natural and popular explanation is that they are due to nodes in the gap arising from unconventional pairing of d-wave type [1], [4], [5], an interpretation also conveyed by SQUID experiments [17], [18]. Our results provide support to d-wave pairing, because they show a linear behaviour in films, but also because they eliminate the uncomfortable need to explain the T^2-dependence by impurity scattering at the unitary limit [12].

The experimental transmission set-up uses carcinotron tubes as powerful, stable microwave sources and oversized waveguides in order to change easily the frequency (120 to 510 GHz) [19], [20]. We took an extreme care to lower microwave leakages down to 60 dB. The transmitted signal is detected by a helium-cooled InSb bolometer. Measurements have been performed by slowly varying the temperature between 5 and 110 K at fixed frequencies. The transmission of the substrate was checked to vary by less than 1% between 5 and 110 K. We have screened the samples from stray magnetic field, so that the residual field is less than 0.5 G. For a film of thickness d (smaller than the skin depth in the normal state or the penetration depth in the superconducting state) deposited onto a substrate of index n, the transmission writes [21]

$$T = \frac{1}{\left| 1 + \frac{\sigma d Z_0}{1+n} \right|^2} \, , \tag{1}$$

where $\sigma = \sigma_1 - i\sigma_2$ is the complex conductivity of the film, Z_0 the impedance of free space and $Z = 1/\sigma d$ the impedance of the film. If in the energy and temperature range of interest $\sigma_2(T) \gg \sigma_1(T)$ and $(\sigma_2 d Z_0)/(1+n) \gg 1$, we can write

$$\frac{T}{T_{110}} = \mu_0^2 \omega^2 \lambda^4(T) \sigma_{110}^2 \, , \tag{2}$$

where σ_{110} is the normal 110 K conductivity of the film. We choose to normalize the transmission at 110 K because the ratio thus obtained is experimentally more reliable than the absolute value of this transmission. For simplicity we assumed $(\sigma_{110} d Z_0)/(1+n) \gg 1$ in (2). Thus, the measurement of T/T_{110} yield an absolute value of $\lambda(T)$. The uncertainties on the $\lambda(T)$ value which arise when neglecting the interferences within the film and/or the substrate, and the effect of the finite film thickness with respect to $\lambda(T)$ have been estimated: the transmission is fairly insensitive to the interference effects at our frequencies as can be shown by comparing a complete expression for T to (1). Moreover, no significant change of the transmitted energy could be observed in the most sensitive range 440–550 GHz [20].

TABLE I. – *Characteristics of the* $YBa_2Cu_3O_7$ *thin films.* d *is their thickness and* $\Delta\theta$ *the rocking curve width.* $R_S(77$ K, 10 GHz) *has been corrected for the film thickness.*

Sample	$d\,(\text{Å})$	$T_c\,(\text{K})$	$\Delta\theta(\text{degrees})$	$R_S(\text{m}\Omega)$	$\sigma_{110}(\mu\Omega\,\text{cm})$
$A1$	1200	86	0.8	~ 10	130
$B1$	1400	88	0.42	0.45	
$B2$	1000	92		0.27	80

The finite film thickness yields an approximate 80 Å overestimate of $\lambda(T)$ for $d/\lambda(T) \sim 0.5$. $\sigma_2(\omega, T) \gg \sigma_1(\omega, T)$ holds at low temperature ($T \leq 20$ K) for all frequencies, but should break down at 40 K [5], [22], [23]. Indeed, σ_1 (35 GHz, 40 K) $\sim 2 \times 10^7 \Omega^{-1}$ m^{-1}, a value likely larger than σ_2 (300 GHz). However, we expect a strong decrease of $\sigma_1(T)$ at 300 GHz [22]-[24].

It is essential that we define the criteria we use in order to sort out the films that we investigate. Our films are epitaxially grown either by laser ablation on MgO [25] or by sputtering on LaAlO$_3$ [26]. They have a narrow transition ($\Delta T_c \leq 1$ K). T_c is 86–92 K, depending on the substrate. In previous papers, the correlation between the film quality in terms of transmission, width of the rocking curve and surface resistance has been established [19], [20], [27]. This led us to select the films either from their surface resistance $R_S \leq 0.5$ mΩ at 77 K and 10 GHz, or the width of their rocking curve $\Delta\theta \leq 0.5°$. The characteristics of our samples are listed in table I. Two types of films have been intentionally investigated. The first film ($A1$) is of poor quality with respect to the above criteria. The other two ($B1$ and $B2$) display either a low surface resistance or a narrow rocking curve. We show in fig. 1 and fig. 2 the normalized transmission of the $A1$ and $B2$ films, respectively. Strong differences
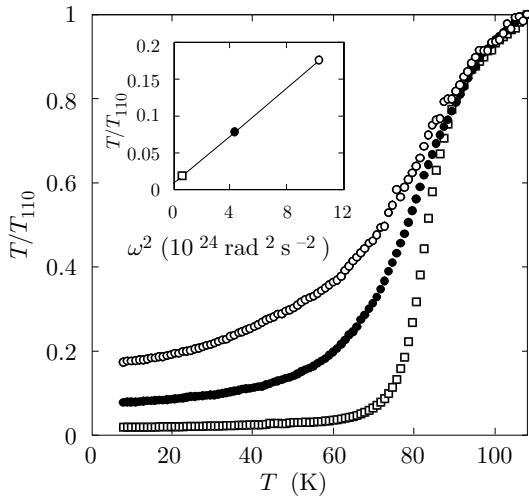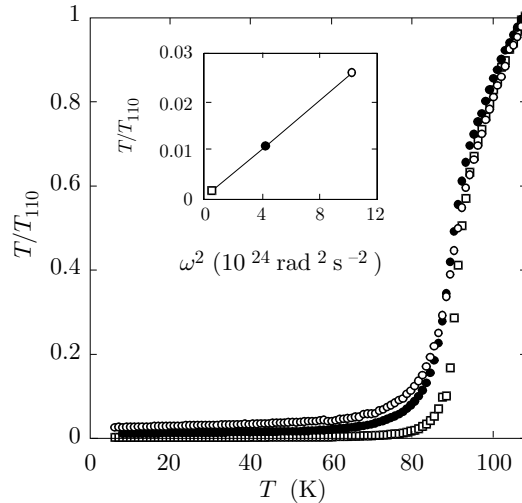

Fig. 1.

Fig. 2.

Fig. 1. – 110 K normalized transmission *vs.* temperature for the YBCO $A1$ film. The inset shows the ω^2 law of the normalized transmission at 10 K. □ 120 GHz, ● 330 GHz, ○ 510 GHz.

Fig. 2. – 110 K normalized transmission *vs.* temperature for the YBCO $B2$ film. The inset shows the ω^2 law of the normalized transmission at 10 K. Same notations as fig. 1.
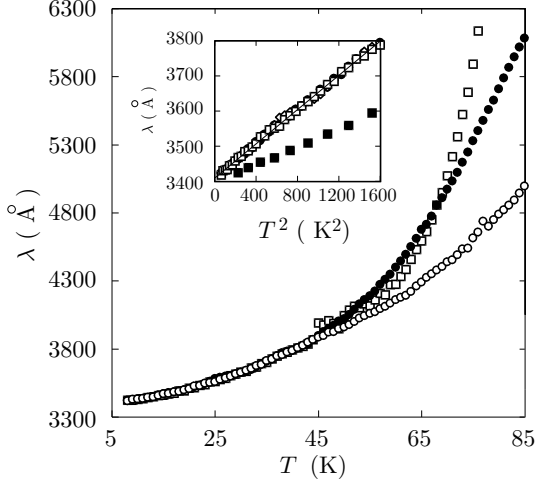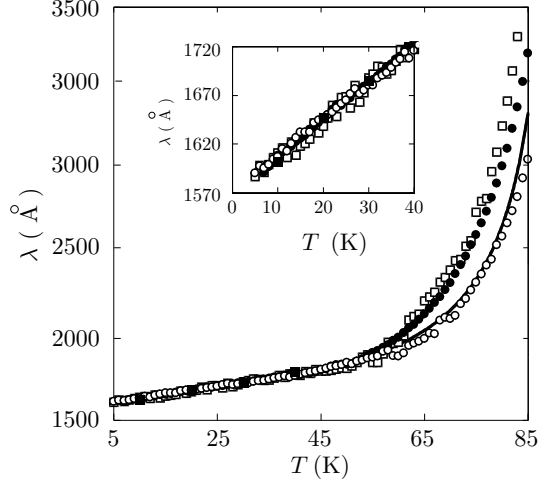
Fig. 3.

Fig. 4.

Fig. 3. – Temperature dependence of $\lambda(T)$ for the YBCO $A1$ film. The T^2 law (see inset) is characteristic of a poor-quality film. The bold squares represent the 8 GHz Porch *et al.* [2] data points shifted to our $\lambda(0)$ value. Same notations as fig. 1.

Fig. 4. – Temperature dependence of $\lambda(T)$ for the YBCO $B2$ film. The bold squares represent the Hardy *et al.* [4] data points on a single crystal (shifted to our $\lambda(0)$ value). The solid line is a comparison with a d-wave–like, strong-coupling calculation, with $2\Delta_0 = 7k_B T_c$ (see text). Same notations as fig. 1.

are observed, in particular for the residual transmission in the superconducting state. At $T \leq 0.5T_c$, the transmission increases as ω^2 in sample $B2$, as expected from (2) and as shown in the inset of fig. 2. In sample $A1$, the data can be analysed by adding a constant T_0 to the ω^2 term (^{2}). We estimate T_0 from the 10 K data (see inset of fig. 1). We compute $\lambda(T)$ from (2) for the three frequencies, however incorporating finite film thickness corrections, assuming that T_0 in the case of $A1$ does not depend on temperature or frequency. The result of this analysis is shown in fig. 3 and fig. 4, for the samples $A1$ and $B2$. The 120, 330 and 510 GHz curves collapse, up to 40 K for sample $A1$ and up to 55 K for sample $B2$. This confirms that the frequency-dependent part of the transmission varies as ω^2, and that $\sigma_2(\omega, T) \gg \sigma_1(\omega, T)$ in these temperature ranges. $\lambda(0)$ is found to be 1570 ± 200 Å for $B2$ and 3400 ± 200 Å for $A1$. The uncertainty on $\lambda(0)$ depends mostly on the accuracy on the σ_{110} measurement, which is limited by the uncertainty on the film thickness (± 100 Å) and by the Van der Pauw technique. Finally, the most striking result is the temperature dependence of $\lambda(T)$. For sample $A1$, we find, as shown in the inset, a clear T^2-dependence, a fairly common result for thin films [1]-[3], associated with a very large value for $\lambda(0)$. In contrast, for sample $B2$, we find a linear temperature dependence up to 50 K along with a much shorter $\lambda(0)$. Such a linear behaviour was similarly observed in film $B1$.

We now discuss the implications of these results. We believe that they demonstrate clearly the extrinsic origin of the T^2-dependence found previously in thin films. Indeed they agree with a phenomenological expression $\lambda^2(T) = \lambda_{\text{intr}}^2(T) + \lambda_{\text{extr}}^2(T)$ as proposed by Hylton *et al.*,

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(^{2}) One possible origin for the extra transmission T_0 could be holes in the film: indeed observations with the optical microscope show in film $A1$ few holes of 100 μm typical size which may give rise to excess transmission.

with λ_{extr} being, for example, the contribution of weak links (but our conclusions are obviously independent of this specific expression). When λ_{extr} is the dominating contribution as in our film $A1$, it may produce the T^2-dependence which is likely to change from sample to sample. Indeed, the data of Porch *et al.* (see inset of fig. 3) are not the same as ours. On the other hand, for good enough films, λ_{extr} gets negligible and we find the intrinsic behaviour for $\lambda(T)$ which is linear, and is expected to be unchanged from films to crystals. The YBCO single-crystals results of Hardy *et al.* are reported in fig. 4, shifted to our $\lambda(0)$ value. The excellent agreement between the two sets of data confirms the intrinsic nature of λ in film $B2$. We remark that the agreement between $\lambda(T)$ in two completely unrelated samples is a very strong result since, except for a coincidence, it eliminates any extrinsic explanation like poor surface effects or pair breaking due to magnetic impurities [10], for the interpretations of the linear dependence. However, the nature of the low-energy excitations is still unknown [28], [29] and it is also not yet clear whether they are linked to the planes or to the chains. Furthermore, the T^2-dependence appears in a film where the penetration depth is large. Our $\lambda_{\mathrm{intr}}(0)$ is much shorter. It is larger though than $\lambda_{ab}(0) = 1450$–1490 Å derived by µSR [30], and consistent with $\lambda_a(0) = 1600$ Å provided by infrared reflectance data [31]. We believe that most techniques do not provide as a straightforward determination as ours.

Our results offer a solution to a previous uncomfortable situation arising in the d-wave interpretation of the experimental results for $\lambda(T)$. Indeed, in order to explain the T^2 behaviour in films together with the linear dependence in single crystals, theoretical calculations had to call for a high impurity concentration in films. But this implies a significant difference between T_c for films and crystals which is not observed. The proposed escape [32] was to assume that the impurity concentration was rather low with a scattering very near the unitary limit, a very specific hypothesis. In this case T_c could be little changed by impurities while the low-T behaviour of $\lambda(T)$ would be much more affected. Since our results show that the T^2 behaviour is extrinsic, *e.g.* due to weak links [20], all the results become coherent with a reasonably low content of ordinary scatterers.

In order to interpret quantitatively our results, it is convenient to make use of the simplified two-dimensional model of d-wave–like pairing introduced by Xu *et al.* [12], where the gap has a linear dependence near the nodes and is constant elsewhere: $|\Delta(\theta)| = \mu\Delta_0\theta$ for $0 \le \theta \le 1/\mu$ and $|\Delta(\theta)| = \Delta_0$ for $1/\mu \le \theta \le \pi/4$, θ being the angle with respect to the node position on the Fermi surface and the rest of $\Delta(\theta)$ being obtained by symmetry. The density of states is taken as constant. This model contains the essential physical ingredients to represent a general order parameter with d-wave symmetry, except for the possible effect of Van Hove singularities. The slope of $\lambda(T)$ at $T = 0$ is given by $2\ln 2/\mu\Delta_0$, however when Δ_0 is calculated with a weak-coupling assumption, the result can only be made to agree with experiment up to 20 K and it does not reproduce the remarkable linear behaviour up to 55 K. A simple way to account for this feature is to incorporate strong-coupling effects. This is justified on theoretical ground [8] and is also consistent with experiments since, for example, tunnelling data give an effective gap larger than what is expected from BCS theory. If we take $2\Delta_0/T_c = 7$ (a typical tunnelling value), we obtain the solid curve in fig. 4 which agrees quite well with our experimental results. This gives further support to this interpretation. In summary, although our results cannot be taken as a proof for d-wave pairing, they put at least this interpretation in a much better position.

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