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Penetration depth in $YBa_2Cu_3O_7$ thin films from far-infrared transmission

L. A. de Vaulchier, J. P. Vieren, A. El Azrak, Y. Guldner, and N. Bontemps
Laboratoire de Physique de la Matière Condensée (URA CNRS 1437), ENS, 24 Rue Lhomond, 75231 Paris Cedex 05, France

M. Guilloux-Viry, C. Le Paven-Thivet, and A. Perrin
Laboratoire de Chimie du Solide et Inorganique Moléculaire (URA CNRS 1495), Université de RENNES I, 35042 Rennes Cedex, France
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We compare the analytical expressions for the transmission and the surface resistance of a thin superconducting film and we point out the differences and similarities between the two quantities. We show that in the case of the transmission, this single quantity allows a straightforward determination of the electromagnetic penetration depth $\lambda(T \ll T_c)$ at low temperatures. We illustrate this point by transmission measurements at different fixed far-infrared frequencies (4–60 cm^{-1}) in two purposely very different $YBa_2Cu_3O_7$ thin films. We find 2900 and 2000 Å, respectivley, for the absolute value of their electromagnetic penetration depth at $T \ll T_c$, which suggests an extrinsic origin of the field penetration in these samples. Such an assignment may further clarify the temperature dependence of the penetration depth which has been observed in thin films.

I. INTRODUCTION

Transmission measurements in the range 2–100 cm^{-1} were initially used to study low-T_c thin superconducting tin and lead films by Glover and Tinkham,[1] as a successful test of BCS theory. There are, to our knowledge, few such measurements on high-temperature superconductors.[2–5] Nevertheless, these materials have been very thoroughly investigated up to ~90 GHz (3 cm^{-1}), essentially by surface impedance measurements using various resonating techniques[6–12] but also with nonresonant techniques, either transmission or absorption.[3–5,13–15] The surface resistance of thin films has been extensively studied in view of microwave applications.

In search of a basic understanding of the pairing mechanism, the surface reactance of single crystals and thin films has been measured, since it allows the determination of the temperature dependence of the penetration depth $\lambda(T)$. This quantity reflects indeed the anisotropy and possibly the existence of nodes in the superconducting gap.[16,17] In high-quality $YBa_2Cu_3O_7$ crystals, $\lambda(T)$ has been found to exhibit a linear temperature dependence up to ~40 K,[18] consistent with the occurrence of nodes in the gap, which may suggest a *d*-wave pairing mechanism. In thin films, on the other hand, the temperature dependence of the penetration depth is often found to be quadratic with temperature.[19–21] Possible reasons for this discrepancy are (i) within the framework of *d*-wave pairing, scattering due to impurities or defects, which are then assumed to be more numerous in thin films and which may conceal the actual temperature dependence of $\lambda(T)$,[17,22] (ii) weak links, more likely to be present in thin films rather than in high-quality single crystals, which may yield an effective penetration depth λ_{eff} whose temperature dependence would be different.[23,24] Note that in the latter case, λ_{eff} should be larger than the intrinsic depth λ_l: this point cannot be established reliably through surface impedance techniques where the quantity which is actually measured is the variation $\Delta\lambda(T)=\lambda(T)-\lambda(0)$, because the geometric factors involved in the determination of $\lambda(0)$ are not known accurately enough.[18,25] Still the knowledge of $\lambda(0)$ would allow us to decide whether one measures an extrinsic value rather than the intrinsic one.[26,27]

The purpose of this paper is to show how the frequency dependence of the transmission can provide the absolute value of the penetration depth in the temperature range $T \ll T_c$, where the imaginary part of the conductivity is larger than the real part. This is illustrated by transmission measurements in thin films at various frequencies. We discuss experimental difficulties, e.g., extra transmission due to weak links, trapped vortices, microwave leakages, interference effects within the substrate. We show that as long as they do not alter the expected quadratic dependence of the transmission versus frequency, the determination of $\lambda(T \ll T_c)$ is still valid. Our technique will be therefore highly suitable of the search of the temperature dependence of the penetration depth, if properly analyzed, but this issue is not addressed in the present paper.

We recall in Sec. II the expressions for the transmission and the surface resistance, in order to stress their similarities and their differences. We introduce the effect of weak links and of vortex dissipation. As long as the transmission is controlled by the imaginary part of the conductivity, which is the case at low temperature, it allows to determine the absolute value of the electromagnetic penetration depth, whether extrinsic (λ_{eff}) or intrinsic (λ_l). By contrast, the surface resistance besides involving $\lambda_{eff}(T)$, is controlled by the residual real part of the conductivity in the superconducting state, which is generally unknown. Section III describes transmission results on two $YBa_2Cu_3O_{7-\delta}$ thin films, in the range 3–60 cm^{-1}. In Sec. IV, we analyze these data: as discussed in Sec. I, although we observe a nonzero residual transmission at low temperature, we may rely on the fre-

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quency dependence of the transmission to extract the value of the effective penetration depth at 25 K. We also briefly discuss this residual low-temperature transmission, which is especially large on one of the samples, and assign it to vortices in weak links.

II. A COMPARISON BETWEEN TRANSMISSION AND SURFACE RESISTANCE

The transmission of a thin film depends on its complex conductivity $\sigma(T)$:

$$\mathbf{T}=\frac{1}{|1+\sigma d Z_0/(1+n)|^2} . \tag{1}$$

d is the thickness of the film, Z_0 is the vacuum impedance, and n the index of the substrate. Equation (1) is the simple expression already used by Glover and Tinkham,[1] which is valid when the film is thin compared to the skin depth (in its normal state) and to the penetration depth (in the superconducting state). The surface impedance then writes $Z=1/\sigma d$. These conditions are fulfilled for the samples that we have examined (respectively, 1000 and 500 Å). Equation (1) neglects the interference pattern within the film and the substrate. We have also used the complete expression including the interferences.[1,28] However, the results on the penetration depth are not significantly affected, as we will conclude later, and for the sake of the discussion, we shall present the data analysis using the simplified expression.

In order to work out a general expression for the transmission, which would account for an intrinsic and extrinsic field penetration, we use the simple weak-link model suggested by Hylton *et al.*, where the film is described by a network of superconducting grains and resistively shunted Josephson junctions.[23] R_j is the unit areal resistance and L_j the unit areal inductance of the weak links, a is the grain size. There appears two limiting cases, corresponding to the junctions being essentially inductive or resistive. These limits may be defined by a characteristic time $\tau_j=L_j/R_j$ being small or large with respect to $1/\omega$. The inductive regime is set by $\omega\tau_j \ll 1$, the resistive one by $\omega\tau_j \gg 1$.

(a) In the limit $\omega\tau_j \ll 1$, one deals with a superconductor which exhibits an effective penetration depth λ_{eff}:

$$\lambda_{\text{eff}}^2=\lambda_l^2+\lambda_j^2 , \tag{2}$$

where λ_l is the London penetration depth and λ_j is the Josephson length defined by

$$\lambda_j=\sqrt{h/4\pi e a J_c \mu_0} . \tag{3}$$

J_c is the critical current density of the junction and a the size of a superconducting grain. At low enough temperature, $T \ll T_c$, as the imaginary part $\sigma_2(T)$ of the intrinsic conductivity becomes much larger than its real part $\sigma_1(T)$, $\sigma_{2\text{eff}}(T)$ is given by

$$\sigma_{2\text{eff}}(T)=\frac{1}{\mu_0\omega\lambda_{\text{eff}}^2(T)} . \tag{4}$$

The condition $\omega\tau_j \ll 1$ sets $\sigma_{2\text{eff}}(T) \gg \sigma_{1\text{eff}}(T)$. One derives a useful simple expression for the ratio $\mathbf{T}(T)/\mathbf{T}_\mathrm{n}(T_0)$, where $\mathbf{T}(T)$ is the transmission at a temperature $T \ll T_c$ and $\mathbf{T}_\mathrm{n}(T_0)$ the transmission at some reference temperature T_0 in the normal state:

$$\mathbf{T}(T)/\mathbf{T}_\mathrm{n}(T_0)=\mu_0^2\omega^2\lambda_{\text{eff}}^4(T)\sigma_n^2(T_0) . \tag{5}$$

$\sigma_n(T_0)$ is the conductivity (real at our operating frequencies) at temperature T_0. Equation (5) is obtained when neglecting $(n+1)/dZ_0$ with respect to $\sigma_n(T_0)$. All the terms will be, however, taken into account in the actual calculation.

It is interesting to compare this expression to the surface resistance R_s established within the same approximation framework, namely $Z=1/\sigma d$, hence $R_s \sim \sigma_1(T)/\sigma_2^2(T)d$ and $R_n=1/\sigma_n(T)d$ (σ_1 and σ_2 stand for the effective conductivities):

$$R_s(T)/R_n(T_0)=\mu_0^2\omega^2\lambda_{\text{eff}}^4(T)\sigma_1(T)\sigma_n(T_0) . \tag{6}$$

Equation (6) looks very similar to Eq. (5), with the noticeable and simplifying difference that the residual normal conductivity in the superconducting state in Eq. (6) is replaced in (5) by the normal-state conductivity at the reference temperature T_0. This comes from the basic fact that the transmission is controlled at low temperature by the inductive response of the superconductor.

(b) In the opposite limit, $\omega\tau_j \gg 1$ (resistive regime), one has to consider two cases.

$\sigma_2 R_j/a \ll 1$, the transmission and the surface resistance are still given by (5) and (6).

$\sigma_2 R_j/a \gg 1$, then

$$\mathbf{T}(T)/\mathbf{T}_\mathrm{n}(T_0)=\left[\frac{R_j}{a}\right]^2\sigma_n^2(T_0)+\mu_0^2\omega^2\lambda_l^4(T)\sigma_n^2(T_0) . \tag{7}$$

The surface resistance is similarly

$$R_s(T)/R_n(T_0)=\frac{R_j}{a}\sigma_n(T)+\mu_0^2\omega^2\lambda_l^4(T)\sigma_1(T)\sigma_n(T_0) . \tag{8}$$

Note that one recovers in both cases the intrinsic contribution, but now as a correction to the larger resistive contribution. This describes obviously the case of very poor grain boundaries.

It is easy to show that similar equations describe any superimposed dissipation mechanism which may be represented by a resistance ρ in series with the superconductor. Therefore a general formula for the transmission can be written as

$$\mathbf{T}(T)/\mathbf{T}_\mathrm{n}(T_0)=\rho^2\sigma_n^2(T_0)+\mu_0^2\omega^2\lambda_{\text{eff}}^4(T)\sigma_n^2(T_0) . \tag{9}$$

The result that we wish to stress is that one can deduce from the ω^2 frequency variation of $\mathbf{T}/\mathbf{T}_\mathrm{n}$ the absolute value of the effective depth at low temperature, irrespective of the unknown conductivity $\sigma_1(T)$ which appears in the surface resistance, irrespective of ill-defined geometrical factors which preclude its determination from the surface reactance, and to some extent, irrespective of spurious transmission mechanisms. The expected information is then that the better the film, the closer λ_{eff} should be to the intrinsic λ_l value, which is a valuable indication if one is to address an intrinsic property.

III. EXPERIMENT

We illustrate now this discussion with transmission data on two thin films labeled A and B:

A is a $YBa_2Cu_3O_7$ laser-ablated c-axis-oriented film on an $\sim 500\ \mu m$ thick $LaAlO_3$ twinned substrate. Its critical temperature T_c is 86 ± 2 K, its residual resistance ratio (RRR) is $R(300\ K)/R(100\ K) \sim 2$. The film thickness is approximately 1000 Å.

B is a $YBa_2Cu_3O_7$ laser-ablated c-axis-oriented film on an $\sim 500\ \mu m$ thick MgO substrate. The critical temperature T_c is 86 ± 0.5 K, the RRR is 2.5 and the film thickness is approximately 500 Å.

From the above characteristics, A is not a high quality film. B is better; its critical temperature is somewhat low, which is not unusual with MgO substrate.[29]

Figure 1 shows a schematic block diagram of the experimental setup. We have designed this setup in view of transmission at low temperature which may be as low as 10^{-6} (by far not achieved in the data reported here). We therefore use an InSb or silicon helium-cooled detector, and the high power sources available in our laboratory: carcinotrons cover the range 4 cm^{-1} (120 GHz) to 17 cm^{-1} (510 GHz) and a far-infrared CO_2 pumped molecular laser which exhibits 6 lines from 39 cm^{-1} (1.17 THz) to 104 cm^{-1} (3.12 THz). In order to change easily the frequency, our setup takes advantage of overdimensioned brass tubes. This precludes a well defined normal incident angle and/or polarization of the waves transmitted through the sample, which may be a limitation since we are interested in the penetration depth λ_{ab} associated with currents within the ab plane. Nevertheless, the contamination by a small λ_c component is negligible considering the aspect ratio of the films (typically $5 \times 5\ mm^2$). The sample is sealed with silver paint on a brass ring which is itself adjusted into the waveguide. The latter is surrounded by a copper block for thermal anchoring. A platinum resistor is placed in the copper block close to the sample.

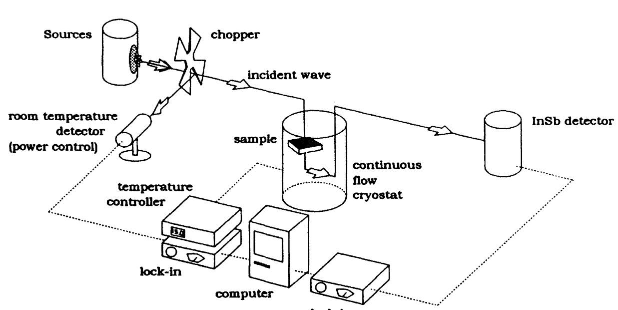

FIG. 1. Block diagram of the experimental transmission setup. The helium-cooled InSb detector is used in the 4–20 cm^{-1} range, the helium-cooled Si detector from 20 to 60 cm^{-1}. The magnetic field (not shown) is vertical when present.

In the set of data reported in this paper, the spurious leakage was estimated to be of the order of 10^{-3} and we consider it constant in the frequency range investigated. The A sample was placed in a superconducting coil, so that a magnetic field could be applied, the counterpart being the remanent field of the coil (~ 50 G). For the B sample, we have used a different cryostat, where the residual field was less than 5 G. The effect of trapped field on transmission is discussed in the last paragraph in relation with an applied field.

We show in Figs. 2(a) and 2(b) the ratio $\mathbf{T}(T)/\mathbf{T}_n(100\ K)$ versus temperature T, at different frequencies, for the A and B samples. We have checked that the bare $LaAlO_3$ substrate transmission changes less than 8% for frequencies below 40 cm^{-1} when the temperature is lowered from 110 down to 10 K but exhibits a larger temperature variation at higher frequencies.[28] The frequency range has therefore been restricted for sample A to the range 2–40 cm^{-1}. The bare MgO substrate transmission has also been checked and is less sensitive to temperature

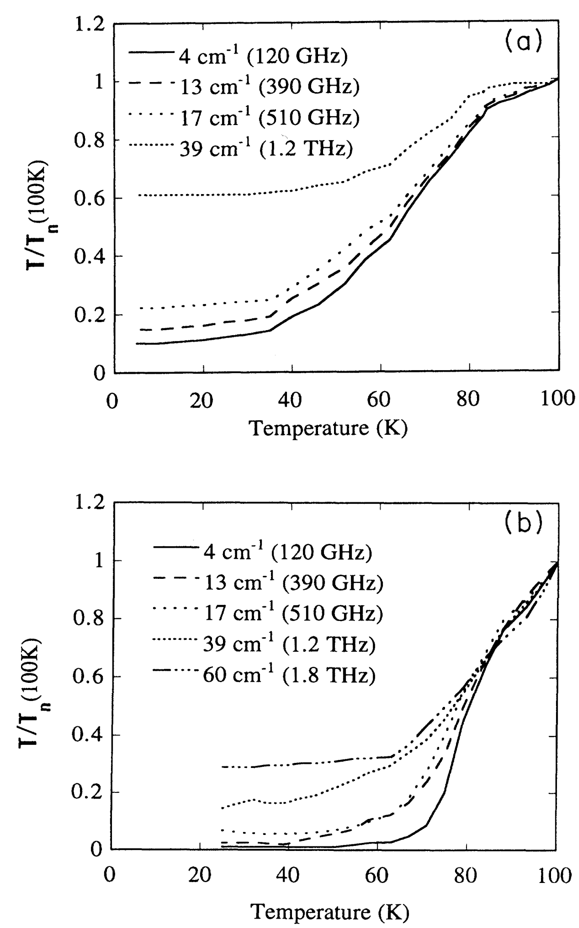

FIG. 2. (a) Transmission ratio $\mathbf{T}(T)/\mathbf{T}_n(100\ K)$ versus temperature at different frequencies for the $YBa_2Cu_3O_7$ A film deposited on the $LaAlO_3$ substrate. (b) Transmission ratio $\mathbf{T}(T)/\mathbf{T}_n(100\ K)$ versus temperature at different frequencies for the $YBa_2Cu_3O_7$ B film deposited on the MgO substrate.

changes, so that one can work up to 60 cm^{-1}.[30] Larger frequencies (above 60 cm^{-1}) may become comparable to the quasiparticle scattering rate, which further complicates the quantitative analysis.[12,29]

IV. DISCUSSION

For both samples, the ratio $\mathbf{T}(T)/\mathbf{T}_n(100\ \mathrm{K})$ decreases significantly as the superconducting state is entered at $\sim$86 K. Note that the temperature range over which $\mathbf{T}(T)/\mathbf{T}_n(100\ \mathrm{K})$ decreases is very large, especially for the A sample. This range broadens as the frequency ω increases, and we therefore had to define a frequency-dependent transition width $\Delta T_c(\nu)$: it is the range over which the transmission drops from 10% down to 90% from its reference normal-state value. We have related $\Delta T_c(\nu)$ to the crystalline quality and c-axis orientation of the films.[32] We have never observed $\Delta T_c(\nu=120\ \mathrm{GHz})<8$ K on the set of films (among which A and B) that we have studied until now.[28]

We now restrict the quantitative analysis to the low-temperature data. Below $\sim$25 K ($\sim$50 K) for sample $A(B)$, $\mathbf{T}(T)/\mathbf{T}_n(100\ \mathrm{K})$ reaches a residual value of 0.1 (0.01) for the A (B) sample. We have reported in Figs. 3(a) and 3(b) the ratio $\mathbf{T}/\mathbf{T}_n(100\ \mathrm{K})$ versus frequency for A and B for such a temperature range. We have to compute the ratio $\mathbf{T}/\mathbf{T}_n(100\ \mathrm{K})$ from Eq. (9) in order to determine $\lambda_{\mathrm{eff}}(T \ll T_c)$. For this purpose, we need the normal-sate conductivity at 100 K. Its precise determination would require a patterning of the samples. We did not do it simply because it prevents further use of the samples for far-infrared measurements. We rely on resistivity measurements which have been performed on thin films exhibiting similar characteristics and we take their value of 120 $\mu\Omega$ cm at 100 K. This fairly high value may be related to weak links, but it is difficult to rule out a possible oxygen deficiency. We also need the indices of the substrate [five for $LaAlO_3$ (Ref. 30) and three for MgO (Ref. 31)], the thickness of the films and of the substrates. Table I shows their nominal values.

We show in Figs. 3(a) and 3(b) the computed curves of the ratio $\mathbf{T}/\mathbf{T}_n(100\ \mathrm{K})$ in the simplified expression (without interferences) and the full expression (with interferences). For our available frequencies, it turns out that using any of these two expressions yields the same values for the penetration depth, considering the error bars. The very sharp features associated with the interference pattern are likely to be smeared experimentally. Each figure displays the case of two values for λ_{eff} which adjust reasonably well the experimental points, including a residual spurious resistivity ρ, reported in Table I.

We find $\lambda_{\mathrm{eff}}=2800-3000$ Å for sample A, and $\lambda_{\mathrm{eff}}=1900-2000$ Å for sample B. It appears for both samples anomalously large if compared to the widely accepted value of 1500 Å. This is of course, not surprisingly, a strong indication that the experimentally determined penetration depth is not the intrinsic one. For the A sample, using the weak-link model, we derive $\lambda_j \sim 2500$ Å, which tells that the temperature variation of λ_{eff}, if studied, is to be controlled by the critical current density J_c of the weak links.[23] The problem is less severe for the B sample, where $\lambda_j \sim 1300$ Å.

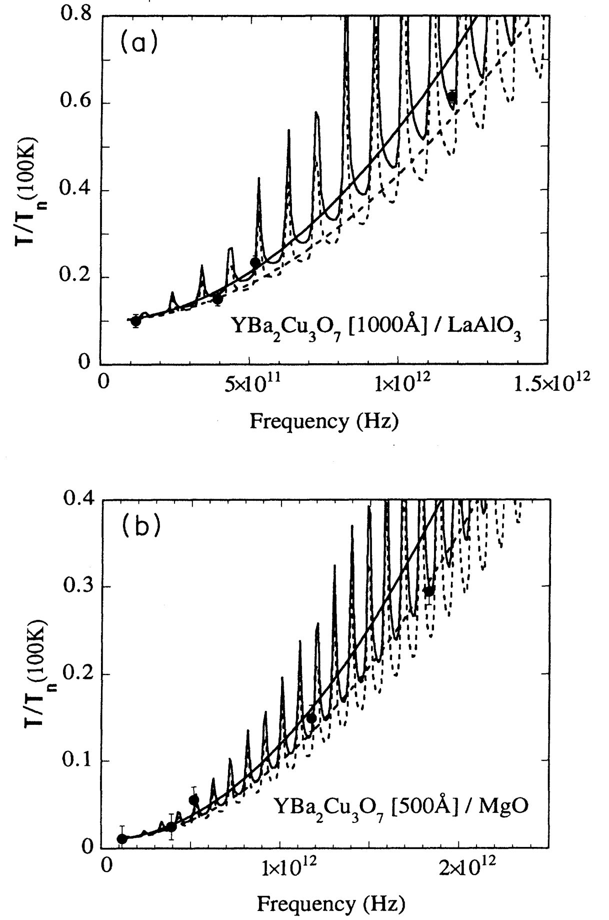

FIG. 3. (a) Frequency dependence of $\mathbf{T}(T)/\mathbf{T}_n(100\ \mathrm{K})$ at $T \le 25$ K for the sample A (full dots) and computed frequency dependence of $\mathbf{T}(0)/\mathbf{T}_n(100\ \mathrm{K})$ with and without interferences in the substrate. The solid lines refer to $\lambda_{\mathrm{eff}}=3000$ Å, the dashed lines to $\lambda_{\mathrm{eff}}=2800$ Å. (b) Frequency dependence of $\mathbf{T}(T)/\mathbf{T}_n(100\ \mathrm{K})$ at $T \le 50$ K for sample B (full dots) and computed frequency dependence of $\mathbf{T}(0)/\mathbf{T}_n(100\ \mathrm{K})$ with and without interferences in the substrate. The solid lines refer to $\lambda_{\mathrm{eff}}=2000$ Å, the dashed lines to $\lambda_{\mathrm{eff}}=1900$ Å.

We briefly discuss the problem of the large transmission on sample A (for the B sample, the residual transmission is negligible compared to the overall varia-

TABLE I. Characteristics of the two samples. λ_{eff} and the low-temperature residual resistivity ρ (see text) are determined from the fit to the computed transmission, d is the film thickness, and e is the thickness of the substrate used in the computation shown in Figs. 3(a) and 3(b).

Sample	$\lambda_{\mathrm{eff}}(T\sim 0\ \mathrm{K})$ (Å)	ρ ($\mu\Omega$ cm)	d (Å)	e (μm)
A	2900±100	35±4	1000	520
B	1950±50	13±4	500	520

tion of the transmission in the frequency range of interest). Such a large transmission at low temperature may be related to the quality of the sample. More precisely, it may be due either to some leakage through the sample itself or to dissipation processes appearing formally as a resistance in series with the superconductor. Therefore one can think of a damaged sample or of a dissipative regime as mentioned in the first part of this paper. Inspection of the film with the resolution of the optical microscope does not reveal, except for the twins of the substrate, any major crack or defect at a few μm scale which might be responsible for major leakage through the film itself. Weak links are ascertained by the large penetration depth: the model of Hylton may not apply to those which are really poor (only resistive). We examine another possibility, namely dissipation due to vortices. Indeed, it was shown that the occurrence of flux flow yields a resistance which appears actually in series with the superconductor resistance.[33] In another study using a parallel plate resonator technique, it was found that for low-quality films, the surface resistance saturates at low magnetic field (0.05 T) and does not depend on the field orientation. In contrast, on good quality films, the increase of the surface resistance is much more gradual on the scale of 1.5 T and no saturation is observed up to this field value.[34] This different behavior was assigned to vortices in weak links inducing a rapid saturation of the surface resistance of thin films with increasing fields. We assume that a similar mechanism is responsible for most of the high residual transmission which is observed, due to trapped field in the film. Indeed, we observed a small increase of the transmission when applying a 12 T field on the A sample: $\mathbf{T}/\mathbf{T}_n$ changes from 0.1 to 0.15, which is not consistent with a quadratic increase of the transmission with field as expected from a flux-flow calculation. It therefore suggests a strong saturation of the losses versus field, consistent with vortices present in weak links already at low field and calls for a careful screening of the samples even from the earth field.[35]

V. CONCLUSION

We have shown that transmission measurements in the range 4–40 cm^{-1} (120 GHz–1.2 THz) in superconducting films yield the absolute value of the electromagnetic penetration depth at low temperature. The values reported for two different films strongly suggest that for those samples, weak links dominate the field penetration. Therefore any temperature variation of the penetration depth is likely to be extrinsic. Such experiments may eventually help to clarify the discrepancies observed between different samples.

The next step to be taken is to establish through the same approach the temperature dependence of the penetration depth. Such measurements are under way in our laboratory and will be discussed and reported elsewhere. We just mention here as an example, that for a laser-ablated $YBa_2Cu_3O_7$ film which exhibits a penetration depth of $\sim$3600 Å, determined in the same way, we find that $\lambda(T)$ follows closely a T^2 temperature behavior from 8 to 45 K.

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