RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 48, NUMBER 17 1 NOVEMBER 1993-I

Temperature dependence of the in-plane penetration depth of $YBa_2Cu_3O_{6.95}$ and $YBa_2(Cu_{0.9985}Zn_{0.0015})_3O_{6.95}$ crystals from T to T^2

D. Achkir and M. Poirier
Centre de Recherche en Physique du Solide, Département de Physique, Université de Sherbrooke, Québec, Canada J1K 2R1

D. A. Bonn, Ruixing Liang, and W. N. Hardy
Department of Physics, University of British Columbia, Vancouver, British Columbia, Canada V6T 1Z1

(Received 9 August 1993)

The low-temperature dependence of the microwave complex surface impedance (17 GHz) of pure and Zn-doped high-quality YBCO single crystals is reported. The surface resistance of the pure crystal shows a peaked structure around 40 K which disappears with Zn doping. The in-plane penetration depth varies linearly with temperature below 25 K; a T^2 contribution appears at the lowest temperature when Zn impurities are added. This suggests that the former temperature dependence is intrinsic to pure crystals whereas the T^2 behavior, also observed in thin films, would be due to defects. The linear temperature dependence is expected for superconducting gaps with lines of nodes.

Since the discovery of high-temperature superconductivity in copper oxides, many experiments have been performed to characterize the nature of this superconductivity, in particular the symmetry of the order parameter. This question continues to be debated for various high-T_c compounds, especially $YBa_2Cu_3O_7$ (YBCO),[1] and also for organic compounds.[2] Among several physical properties, the magnetic penetration depth $\lambda(T)$ is a fundamental quantity which can provide information regarding the symmetry of the electron-pairing state leading to superconductivity; it can reveal, for example, the presence of nodes in the pair-wave function for certain directions in k space. Different techniques can be used to measure this penetration depth: muon spin relaxation (μSR),[3] magnetization,[4] and microwave surface impedance.[5,6] Thin films and single crystals of YBCO compounds have been extensively studied by these techniques; from some experiments, an exponential temperature variation,[5,7] as predicted by the BCS theory, has been reported, in contrast with a power-law temperature dependence at low temperatures measured by others.[6,8] The observation of a power law at low temperatures on $\lambda(T)$ is fundamental to establishing whether the order parameter has nodes.[9] However, we have to keep in mind that the presence of defects in these materials may modify the power law; for example, in thin films, it has been suggested that the T^2 law observed for the in-plane penetration depth may not be intrinsic but instead may be related to the presence of defects.[10] In the present paper, we provide experimental proof that impurities can indeed change the temperature dependence of λ_{ab} from T to T^2. This linear dependence of λ_{ab} is measured without contamination from λ_c and with a simultaneous measurement of the real part of the surface impedance. In order to have direct access to the intrinsic behavior of $\lambda(T)$, one has to use high-quality single crystals; this has been exemplified by microwave experiments of Hardy *et al.*[6] and Bonn *et al.*[11] In high-quality single crystals, the surface resistance shows a broad peak well below T_c (Ref. 12) whose physical origin is attributed to a rapid increase of the scattering time of the normal carriers concomitant with a decrease of the normal fluid density in the superconducting state. This maximum progressively disappears as zinc impurities are added in the structure. The apparently very small surface resistance observed in thin films could then be an indication of the presence of defects, which would result in a T^2 temperature dependence for $\lambda(T)$.[3,5,9,13,14] Hardy *et al.*[6] have measured the in-plane penetration depth $\lambda_{ab}(T)$ on a high-quality YBCO single crystal with a split-ring-type resonator technique[6] and a linear temperature dependence was then observed. Such a dependence can occur for example in a clean d-wave superconductor with line nodes in the gap. There is however some controversy about these data concerning the exact nature of the deduced $\lambda_{ab}(T)$. Since the application of the ac magnetic field was parallel to the ab plane, the observed λ cannot be directly related to the in-plane $\lambda_{ab}(T)$; corrections attributed to $\lambda_c(T)$ have to be included. In the experiment of Hardy *et al.*[6] these corrections should be small, but in order to eliminate any remaining doubts and to confirm the linear dependence at very low temperatures, it is essential to measure directly the temperature dependence of λ_{ab} in high-quality single crystals. We thus report in this paper very precise microwave surface impedance measurements on pure and zinc-doped (0.15 %) YBCO single crystals. We used a conventional perturbation technique which gives simultaneous access to the surface resistance and to the penetration depth. The crystal quality is partly confirmed by the observation of a peak in the surface resistance well below T_c which disappears in the noise for the doped sample. The measured $\lambda_{ab}(T)$ in the pure crystal is in perfect agreement with the linear temperature dependence reported by Hardy *et al.*;[6] this gives additional evidence for an unconventional type of superconductivity in these materials. In the doped crystal we observed a linear temperature dependence with a crossover to a T^2 law at the very lowest temperatures. This last behavior is consistent with the temperature dependence observed on thin films.

The high-quality crystals used in the measurements were grown by a flux method in zirconia crucibles as described elsewhere.[15] The pure crystal $YBa_2Cu_3O_{6.95}$ has a $T_c \simeq 93.7$ K and very sharp transitions in resistivity, magnetization, and specific heat $\Delta T_c < 0.25$ K. The doped crystal $YBa_2(Cu_{1-x}Zn_x)_3O_{6.95}$ with $x = 0.0015$ has been grown by the same technique as that used to produce high purity crystals; but with Zn incorporated into the flux; the transition width is similar to the pure sample but the transition temperature is lower: 92.4 K. These crystals showed no surface texture or defects and are also free of the potential sources of extrinsic loss in films, e.g., grain boundaries, residual flux, and impurity phases. Both types of crystals are twinned and, since the microwave loss is very sensitive to mechanical damage, no effort at detwinning has been made.

A conventional cavity perturbation method is used to measure, with high sensitivity (10^{-6}), the frequency shift $\Delta\omega/\omega$ and the variation of the quality factor $\Delta(1/2Q)$ following sample insertion as a function of temperature at 17 GHz in a TE_{102} transmission mode. The platelet-shaped sample (ab plane) has a 1.3×1.2 mm^2 surface and it was glued to a quartz rod that passes through the cavity; it could then be easily inserted and oriented with the maximum ac field H_{ac} perpendicular to the highly conducting ab plane. In this field configuration the currents are flowing in the plane and the in-plane surface impedance is obtained.

In the skin-depth approximation, the surface impedance $Z_s = R_s + iX_s$ is related to the experimental parameters by the expression[2]

$$\Delta(1/2Q) + i\left[\frac{\alpha}{1-N} - \frac{\Delta\omega}{\omega}\right] = \beta(R_s + iX_s), \qquad (1)$$

where N and α are the depolarization and filling factors, respectively; the constant β is a geometrical factor reflecting the dimensions of the sample and cavity. R_s and X_s are, respectively, the surface resistance and the surface reactance. In terms of the complex conductivity $\sigma^* = \sigma_1 - i\sigma_2$, the surface impedance is given by the equation

$$Z_s = R_s + iX_s = [i\omega\mu_0/(\sigma_1 - i\sigma_2)]^{1/2}. \qquad (2)$$

In the superconducting state ($T < T_c$) the surface reactance X_s is directly connected to the in-plane magnetic penetration depth, namely

$$X_s(T) = \omega\mu_0\lambda_{ab}(T). \qquad (3)$$

where μ_0 is the permeability of free space and ω the angular frequency. The two factors, β and $\alpha/[1-N]$, are difficult to evaluate and this is why the absolute values of the surface impedance were not determined.

In Fig. 1, we display both measurements of the surface resistance $R_s(T)$ and the surface reactance $X_s(T)$ for the pure sample for temperatures in the range 77–115 K. The width of the transition observed on R_s is very narrow, $\Delta T_c \simeq 0.25$ K, and the transition temperature is high, $T_c = 93.7$ K. These parameters are consistent with the values obtained from dc resistance for crystals of the same batch. These parameters are important indications of the high quality of the sample. Above T_c, $R_s(T)$ and $X_s(T)$ are identical, an observation which is compatible with the fact that for $T > T_c$, in the normal skin-depth regime, the real part of the conductivity σ_1 is much larger than the imaginary part σ_2 so that, following Eq. (2), $R_s = X_s = (\mu_0\omega/2\sigma_n)^{1/2}$. The index n refers to the normal-state conductivity. The microwave loss R_s is very small below 90 K and almost saturates to zero on this scale; the surface reactance $X_s(T)$ decreases more slowly with decreasing temperature. For the doped sample, the data (not shown) are identical over this temperature range, with the exception that the critical temperature has been shifted to 92.4 K. These observations agree with the measurements of Zhang *et al.*[12]

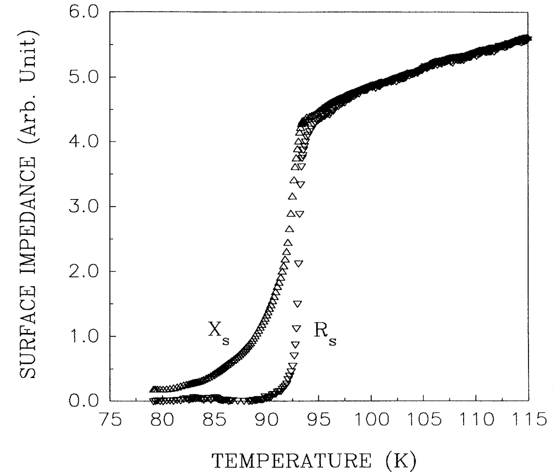

FIG. 1. Surface resistance R_s and surface reactance X_s as a function of temperature for the pure crystal.

Turning our attention to the data at lower temperatures, Figs. 2(a) and 2(b) show $R_s(T)$ for, respectively, the pure and the doped crystals below 60 K. In Fig. 2(a), the surface resistance for the pure crystal presents a broad peak centered around 40 K. This behavior had also been observed by Zhang *et al.*[12] on the same crystal at two different frequencies 3.8 and 35 GHz. Our data obtained at 17 GHz are thus consistent with previous observations of the maximum. In Fig. 2(b), $R_s(T)$ for the doped crystal shows only noise with an average monotonic temperature dependence without the appearance of a peak: this is again consistent with the data of Zhang *et al.*,[12] who observed that the peak is rapidly suppressed by the addition of 0.15 % zinc impurities. In our experiment the signal to noise ratio is not sufficient to confirm the existence of the small plateau observed by Zhang *et al.* for this Zn concentration.[12] Nevertheless, the lower-temperature features observed by Zhang *et al.*[16] are fully consistent with our data and we conclude that our microwave experiment is adequate to study the complete surface impedance of these crystals, including the surface reactance that we now discuss.

The surface reactance $X_s(T)$ for the pure sample,

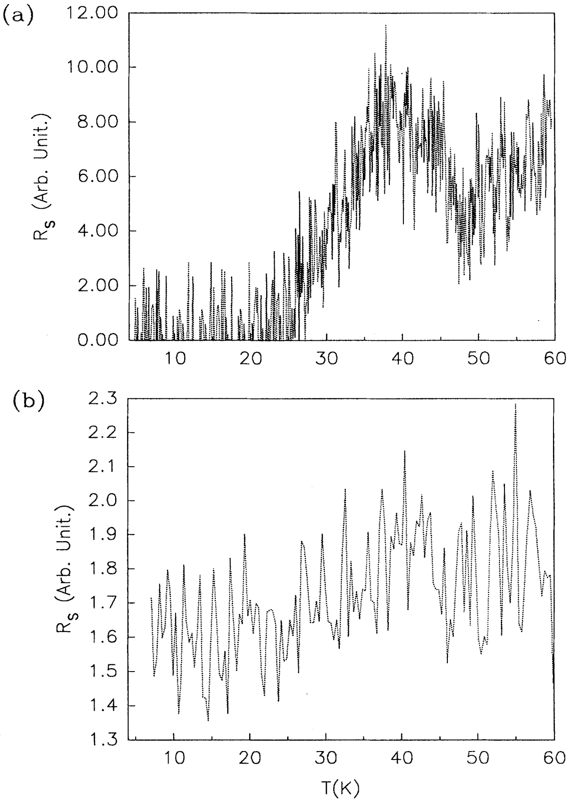

FIG. 2. The temperature dependence of the surface resistance of (a) the pure and (b) Zn-doped crystals.

presents a linear temperature dependence below 25 K, reflecting directly the fact that the in-plane penetration depth depends linearly on temperature. To make a comparison between our results and those of Hardy *et al.*,[6] we fix the absolute scale of our measurements, by requiring that our linearly extrapolated value of $X_s(T)$ at $T = 0$ K equals the $T = 0$ K value of $X_s(0)$ calculated from Eq. (3) using the same value of $\lambda_{ab}(0)$ as Hardy *et al.*[6] This value is $\lambda_{ab}(0) = 1480$ Å as given by the high-field μSR data obtained for crystals of the same batch.[3] Using this procedure to fix the scale, we can now plot

$$\Delta\lambda_{ab}(T) = \lambda_{ab}(T) - \lambda_{ab}(0). \qquad (4)$$

In Fig. 3(a) results for $\Delta\lambda_{ab}(T)$ obtained for the pure sample are displayed on a logarithmic scale for further comparison with the doped sample; the noise is important since we have reached the resolution limit of the experimental technique. However a clear linear temperature dependence is observed below 25 K with a slope of 4.1 Å/K. This is again in full agreement with Hardy *et al.*[6] and it confirms that their observed behavior is really related to $\lambda_{ab}(T)$.

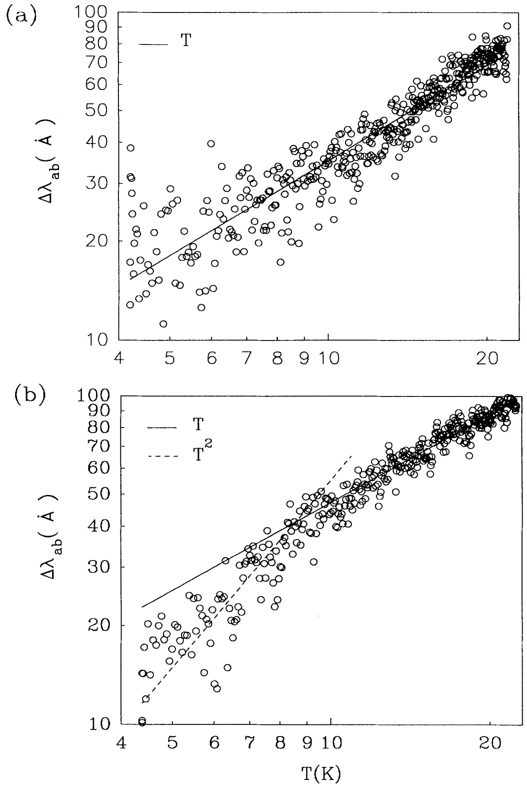

FIG. 3. Variation of the in-plane penetration depth $\Delta\lambda_{ab}$ as a function of temperature on a log-log scale: (a) pure crystal : the solid line represents a linear temperature dependence. (b) Zn-doped crystal : the solid and dashed lines represent, respectively, a linear and a quadratic temperature dependence.

The surface reactance $X_s(T)$ for the doped crystal also presents a linear temperature behavior in the range $10 - 25$ K but a different power law is observed below. These data have been transformed into $\Delta\lambda_{ab}(T)$ by extrapolating the high-temperature ($10 - 25$ K) linear behavior of $X_s(T)$ to zero temperature in the same manner as in the pure sample. In Fig. 3(b) we show the penetration depth on logarithmic scale to better appreciate the power-law dependence at low T. This measurement of $\Delta\lambda_{ab}(T)$ shows a linear variation over the range $10 - 25$ K and the appearance below 10 K of a T^2 law. Recently Arberg *et al.*[10] have calculated the temperature dependence of the London penetration depth for a *d*-wave superconductor and they found that impurity scattering modifies the temperature variation of $\Delta\lambda$ from a T to a T^2 power law beginning at the lowest temperatures. More recently Hirschfeld and Goldenfeld[17] have also calculated the effect of strong impurity scattering on the low-temperature penetration depth of a *d*-wave superconductor and they found a crossover temperature quantitatively consistent with our data. These results would explain why, in YBCO thin films, where the concentration of defects is important, a T^2 law is generally encountered for the penetration depth.

In conclusion this paper reports both real and imaginary parts of the microwave surface impedance obtained during the same experiment on pure and doped high-quality single crystals. Our data clearly demonstrate that the addition of Zn impurities alters drastically both the surface resistance and reactance below T_c. Our results also confirm, on the one hand, previous studies concerning the importance of the rapidly increasing quasiparticle scattering time in the superconducting state and its reduction when defects are present and, on the other hand, the linear temperature dependence of the in-plane penetration depth at low temperatures when the effect of impurities is negligible. The quadratic temperature dependence would then appear at the lowest temperatures when impurity scattering becomes important as suggested by the crossover from T to T^2 observed in our Zn-doped sample. These data give additional evidence for an unconventional type of superconductivity in high-T_c compounds: the linearity with temperature for the in-plane penetration depth is consistent with lines of nodes in the gap function in general, and in particular with d-wave superconductivity.

We acknowledge stimulating discussions with A.-M. Tremblay. We also thank M. Castonguay for technical assistance. This work was supported by Natural Sciences and Engineering Research Council of Canada, by the Canadian Institute for Advanced Research, and by le Fonds pour la Formation de Chercheurs et l'Aide à la Recherche from the Government of Québec.

---

[1] D. A. Bonn, P. Dosanjh, R. Liang, and W. N. Hardy, Phys. Rev. Lett. **68**, 2390 (1992).

[2] D. Achkir, M. Poirier, C. Bourbonnais, G. Quirion, C. Lenoir, P. Batail, and D. Jerome, Phys. Rev. B **47**, 11595 (1993).

[3] D. A. Bonn, Ruixing Liang, T. M. Riseman, D. J. Baar, D. C. Morgan, Kuan Zhan, P. Dosanjh, T.L. Duty, A. MacFarlane, G. D. Morris, J.H. Brewer, W. N. Hardy, C. Kallin, and A. J. Berlinsky, Phys. Rev. B **47**, 11314 (1993).

[4] L. Kruisin-Elbaum, R. L. Greene, F. Holtzberg, A. P. Malozemoff, and Y. Yeshurun, Phys. Rev Lett. **62**, 217 (1989).

[5] J. M. Pond, K. R. Carroll, J. S. Horwitz, D. B. Chrisey, M. S. Osofsky, and V. C. Cestone, Appl. Phys. Lett. **59**, 3033 (1991).

[6] W. N. Hardy, D. A. Bonn, D. C. Morgan, Ruixing Liang, and Kuan Zhang, Phys. Rev. Lett. **70**, 3999 (1993).

[7] S. Sridhar, Dong-Ho Wu, and W. Kennedy, Phys. Rev. Lett. **63**, 1873 (1989).

[8] F. Gao, J. K. Kruse, C. E. Platt, M. Feng, and M. V. Klein (unpublished).

[9] J. Annett, N. Goldenfeld, and S. R. Renn, Phys. Rev. B **43**, 2778 (1991).

[10] P. Arberg, M. Mansor, and J. P. Carbotte, Solid State Commun. **86**, 671 (1993).

[11] D. A. Bonn, Kuan Zhang, D. J. Baar, D. C. Morgan, and W. N. Hardy (unpublished).

[12] Kuan Zhang, D. A. Bonn, Ruixing Liang, D. J. Baar, and W. N. Hardy (unpublished).

[13] Steven M. Anlage and Dong-Ho Wu, J. Supercon. **5**, 395 (1992).

[14] Ju Young Lee and Thomas R. Lemberger (unpublished).

[15] Ruixing Liang, P. Dosanjh, D. A. Bonn, D. J. Baar, J. F. Carolan, and W. N. Hardy, Physica C **195**, 51 (1992).

[16] Kuan Zhang, D. A. Bonn, Ruixing Liang, D. J. Baar, and W. N. Hardy (unpublished).

[17] Peter J. Hirschfeld and Nigel Goldenfeld (unpublished).