Title
Residual Losses in Epitaxial Thin Films of YBa{sub 2}Cu{sub 3}O{sub 7}

Journal
Applied physics letters, 59(18)

Authors
Miller, D.
Richards, P.L.
Etemad, S.
et al.

Publication Date
1991-06-01

LBL-30918
Preprint

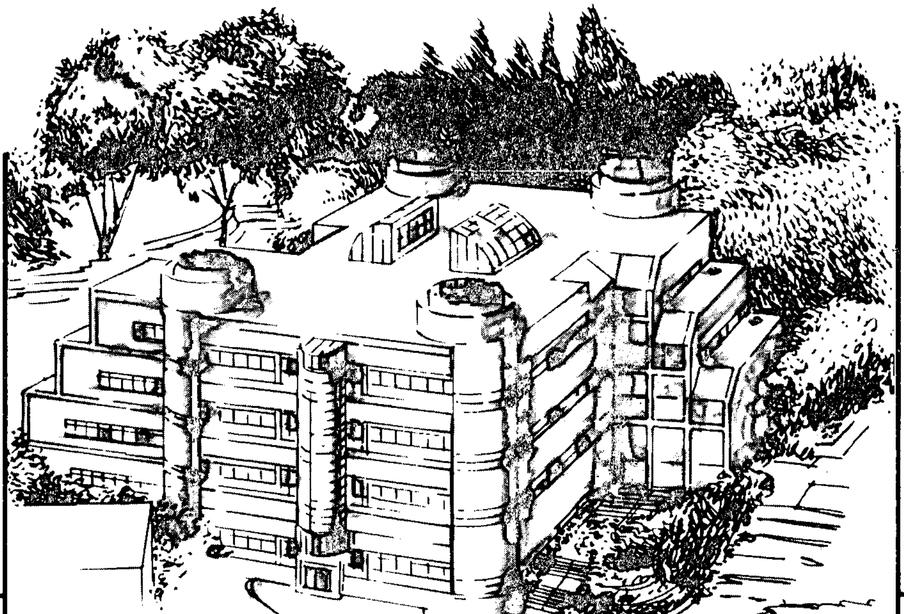
Center for Advanced Materials
CAM

Submitted to Applied Physics Letters

Residual Losses in Epitaxial Thin Films of $YBa_2Cu_3O_7$ from Microwave to Submillimeter Wave Frequencies

D. Miller, P.L. Richards, S. Etemad, A. Inam, T. Venkatesan,
B. Dutta, X.D. Wu, C.B. Eom, T.H. Geballe,
N. Newman, and B.F. Cole

June 1991



Materials and Chemical Sciences Division
Lawrence Berkeley Laboratory • University of California
ONE CYCLOTRON ROAD, BERKELEY, CA 94720 • (415) 486-4755

Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

LBL-30918

Residual Losses in Epitaxial Thin Films of $YBa_2Cu_3O_7$ from Microwave to Submillimeter Wave Frequencies

D. MILLER, P.L. RICHARDS
Department of Physics, University of California, and Materials Sciences Division,
Lawrence Berkeley Laboratory,Berkeley CA 94720

S. ETEMAD, A. INAM, T. VENKATESAN, B. DUTTA, X.D. WU
Bell Communications Research, Red Bank, New Jersey 07701

C.B. EOM, T.H. GEBALLE
Department of Applied Physics, Stanford University, Stanford CA 94305

N. NEWMAN, B.F. COLE
Conductus, Inc., Sunnyvale, CA 94086

PACS Nos. 74.30.Gn, 74.70.Vy, 74.75.+t, 07.62.+s

We gratefully acknowledge C.A. Mears, T.W. Kenny, A.M. Portis, R.C. Taber, J. Beeman, A.T. Barfknecht, W.N. Creager, J.R. Waldram, J. Orenstein and A.E. Zettl for their assistance in various aspects of this work. This work was supported in part by Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U.S. Department of Energy under contract No. DE-AC03-76SF00098, by the AFOSR under contract No. F49620-88-C-004, and by the Center for Research in Superconductivity and Superconducting Electronics under contract No. F49620-88-C-001.

Residual Losses in Epitaxial Thin Films of $YBa_2Cu_3O_7$ from Microwave to Submillimeter Wave Frequencies

D. MILLER, P.L. RICHARDS
Department of Physics, University of California, and Materials Sciences Division, Lawrence Berkeley Laboratory,Berkeley CA 94720

S. ETEMAD, A. INAM, T. VENKATESAN, B. DUTTA, X.D. WU
Bell Communications Research, Red Bank, New Jersey 07701

C.B. EOM, T.H. GEBALLE
Department of Applied Physics, Stanford University, Stanford CA 94305

N. NEWMAN, B.F. COLE
Conductus, Inc., Sunnyvale, CA 94086

PACS Nos. 74.30.Gn, 74.70.Vy, 74.75.+t, 07.62.+s

Abstract

We have measured the residual loss in five epitaxial a-b plane films of the high-T_c superconductor $YBa_2Cu_3O_7$. Microwave measurements near 10 GHz were made by resonance techniques at 4K. Submillimeter measurements from ~1.5 to 21 THz were made at 2K by a direct absorption technique. We use a model of weakly coupled superconducting grains and a homogeneous two-fluid model to fit the data for each film below the well-known absorption edge at 13.5 THz. When the penetration depth determined from muon spin rotation measurements is used to constrain each model, the weakly coupled grain model is able to fit the measured absorptivities for all films, but the two-fluid model is less successful.

The losses in films of high-T_c superconductors such as $YBa_2Cu_3O_7$ (YBCO) at microwave, millimeter and submillimeter frequencies impact many potential applications. Microwave measurements show a temperature dependent component to the loss similar to that for a BCS superconductor plus a substantial residual loss that remains at low temperatures. This residual loss is minimized in high quality epitaxial a-b plane films. Both contributions to the loss vary as frequency squared from ~10 to ~100 GHz. Reflectivity measurements at submillimeter wavelengths show that the losses increase slowly with frequency up to an absorption edge at 13.5 THz (450 cm^{-1}). In order to improve the accuracy of the submillimeter data we have developed a novel technique for directly measuring this residual loss.[1] The high-T_c film is used as absorber in a composite bolometric detector for the signal from a Fourier transform spectrometer with a Hg arc source. The data are normalized by comparison with reference detectors with normal metal absorbers whose absorptivities can be calculated. The residual losses in the films used for this study were also measured near 10 GHz using microwave cavity techniques, and are among the lowest reported in the literature.

The a-b plane samples used in this study were grown on MgO and $LaAlO_3$ substrates, as summarized in Table I. Samples A, B, C and E were produced by 90° off - axis sputtering,[2,3] and film D was produced by pulsed-excimer-laser deposition.[4,5] The samples are notable for their lack of impurity phase and high degree of epitaxial alignment. Film E was intentionally sputtered in an oxygen deficient atmosphere to give a relatively large residual microwave loss. The techniques used to produce the other films were optimized for high film quality.

The results of our measurements are shown in Fig. 1 along with two theoretical fits described below. The curves are displaced by factors of ten to avoid overlap. The filled circles give the microwave loss measurements at 4K. The size of the circles is large enough to include estimated errors and corrections for film transparency. The solid lines give the submillimeter absorptivity measurements at 2K. Additive errors are thought to be small down to the lowest frequencies presented. Multiplicative errors could be as large as 15 percent.[1] These results give a picture of the residual loss in epitaxial a-b plane YBCO films over nearly four orders of magnitude in frequency. In each case, the 10 GHz point can be connected to the submillimeter data by a line which varies as frequency squared. This agrees with observations by others[6] in the range from 10 to ~100 GHz (0.3-30 cm^{-1}). The frequency dependence of the loss saturates smoothly above ~600 GHz (20 cm^{-1}) in a way that is different for different films. There is no sign of any gap-like onset of absorption near 2.1 kT_c as reported in some early work [7] or near the BCS value of 3.5 kT_c = 6.6 THz (220 cm^{-1}). There is a sharp onset of absorption at 13.5 THz (450 cm^{-1}) or 7.2 kTc in our high quality films that has been seen by others in reflectivity experiments.[8,9] Although it seems not to be a BCS-like energy gap, there is clearly a very sudden onset of some kind of new physics. Other investigators have explored the relationship between the residual microwave loss and the submillimeter absorptivity deduced from reflectivity measurements. Our data are generally consistent with early work,[10] but not with a recent preprint which shows higher microwave losses relative to the infrared loss than we observe.[11]

We have used two models of residual loss to fit our data below the absorption edge at 13.5 THz. The first is the model proposed by Hylton *et al.*[12,13] that treats polycrystalline high-T_c films as networks of weakly coupled grains, where the material of the grains is an ideal BCS superconductor. The grain boundaries form resistively shunted Josephson junctions with critical current density j_c, shunt resistivity ρ_J and equivalent Josephson penetration depth λ_j. The grains have characteristic size *a* and superconducting penetration depth λ_g. The magnetic field profile averaged over individual grains is given by $\lambda_{eff} = (\lambda_g^2 + \lambda_j^2)^{1/2}$. This model predicts a frequency squared dependence of the losses at low frequencies and frequency independent losses at high frequencies. The data below 13.5 THz do not contain enough information to uniquely determine the three independent parameters in this model. Consequently, we use the penetration depth obtained from muon spin rotation (μsr) experiments[14] to constrain the fit. This procedure is complicated by the interpretation of the μsr data for inhomogeneous materials. In principle, μsr measures the volume-weighted distribution of magnetic fields throughout the sample. There is a tendency to focus on the rather narrow distribution of fields in the grains which comprise most of the sample volume rather than the broader distribution of fields in the grain boundaries which comprise a much smaller volume. To the extent that this is the case, the μsr experiment measures λ_g, and not λ_{eff}. The fitting is done by characterizing the submillimeter data below 13.5 THz by 20 points equally spaced in log frequency and the microwave data by one point. The two remaining free parameters are then determined by a chi squared minimization technique.

The fits to the weakly coupled grain model, constrained by the μsr measurement[14] with $\lambda_g = 140$ nm, are shown in Fig. 1 as the long dashed lines. The weakly coupled grain model is able to fit the data for all five films measured. The values of λ_j and ρ_J obtained from the fit are given in Table II. Using the fitted parameters, the product of the critical current times the resistance $I_cR = \hbar c^2 \rho_J / 8\pi\lambda_j^2 e$ ranges from 3 to 11 mV for the grain boundaries in our films. This can be compared with a range of 0.2 to 8 mV measured for grain boundary junctions at 4.2K.[15] If we assume that the critical current density j_c of the grain boundaries is essentially the critical current density 3×10^7 A cm^{-2} of the films,[16] then the characteristic grain size *a* = $I_cR/\rho_J j_c$ ranges from 4 to 40 nm in our films. This range is roughly consistent with the spatial correlation range in the laser ablated films of ~ 10 nm,[5] and with twin domain sizes in off-axis sputtered films of ~ 20-200 nm.[16] These values of *a* are considerably smaller than the typical 0.5 to 10 μm size range of the 45° grains in off-axis sputtered films on MgO.[16] Although such high angle grain boundaries are known to behave as resistively shunted junctions[15] which could therefore justify the use of the weakly coupled grain model, the volume fraction of such grains in our films is typically less than one percent.[16,17] Nevertheless, the parameter values deduced from fitting the weakly coupled grain model are reasonable considering the simplicity of the model.

Because the interpretation of the μsr experiments in terms of λ_g is somewhat ambiguous, we have also studied the fit obtained when we constrain λ_{eff} = 140 nm. The best fits shown in Fig. 1 as short dashed lines are acceptable for some, but not all of our films. In the case of film E which has high residual loss, the fit does not even intersect the data. The fit can be forced to intersect the data, but the resulting slope at high frequencies is then so small that the value of chi squared is degraded.

We have also used a type of homogeneous two-fluid model to fit the data which assumes that a fraction of the conduction electrons does not condense into the superconducting state, even at our low temperatures. This model yields a fitting function with the same mathematical form as the weakly coupled grain model. The fitting parameters are taken to be the dc resistivity ρ_o of the uncondensed electrons, their relaxation frequency $\hbar/\tau$ and the penetration depth λ_{tf}, which depends on the density of condensed electrons. When we set λ_{tf} = 140 nm, we get exactly the same unsatisfactory fit (shown by the short dashed lines in Fig. 1) as when we set λ_{eff} = 140 nm in the weakly coupled grain model. The values of ρ_o and $\hbar/\tau$ obtained from this fit are shown in Table II. Our procedure explicitly assumes that the new physics above 13.5 THz does not contribute any oscillator strength below that frequency. In order to test the sensitivity of our two-fluid model parameters to this assumption, we have analyzed our data including oscillator strength at higher frequencies proposed by other investigators.[18,19] The relaxation frequencies obtained are all within the experimental uncertainties reported in Table II.

We have characterized the residual loss in epitaxial a-b plane films of YBCO from 10 GHz to 21 THz. We have shown that the frequency dependence of the loss below 13.5 THz in five films can be well represented by the weakly coupled grain model if the penetration depth in the grains λ_g is set equal to 140 nm. These losses do not seriously affect the performance of passive high T_c microwave devices operated at 77K where the temperature dependent loss is dominant. Experience with a wide range of low T_c devices, however, suggests that many high T_c applications will occur at or below $T_c/2$. The measured residual losses may then seriously affect device efficiency.

We gratefully acknowledge C.A. Mears, T.W. Kenny, A.M. Portis, R.C. Taber, J. Beeman, A.T. Barfknecht, W.N. Creager, J.R. Waldram, J. Orenstein and A.E. Zettl for their assistance in various aspects of this work. This work was supported in part by Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U.S. Department of Energy under contract No. DE-AC03-76SF00098, by the AFOSR under contract No. F49620-88-C-004, and by the Center for Research in Superconductivity and Superconducting Electronics under contract No. F49620-88-C-001.

References

1. D. Miller, P.L. Richards, S. Etemad, T. Venkatesan, L. Nazar, B. Dutta, X.D. Wu, A. Inam, C.B. Eom, T.H. Geballe, N. Newman and B.F. Cole, (submitted to Phys. Rev. B)
2. N. Newman, K. Char, S.M. Garrison, R.W. Barton, R.C. Taber, C.B. Eom, T.H. Geballe and B. Wilkens, Appl. Phys. Lett. **57**, 520 (1990)
3. C.B. Eom, J.Z. Sun, K. Yamamoto, A.F. Marshall, K.E. Luther and T.H. Geballe, Appl. Phys. Lett. **55**, 595 (1989)
4. A. Inam, M.S. Hegde, X.D. Wu, T. Venkatesan, P. England, P.F. Miceli, E.W. Chase, C.C. Chang, J.M. Tarascon and J.B. Wachtman, Appl. Phys. Lett. **53**, 908 (1988)
5. D.M. Hwang, T. Venkatesan, C.C. Chang, L. Nazar, X.D. Wu, A. Inam and M.S. Hegde, Applied Physics Letters **54**, 1702 (1989)
6. H. Piel and G. Muller, IEEE Trans. Magn. **27**, 854 (1991)
7. J. Schutzmann, W. Ose, J. Keller, K.F. Renk, B. Roas, L. Schultz and G. Saemann-Ischenko, Europhys. Lett. **8**, 679 (1989)
8. B. Batlogg, in High Temperature Superconductivity Proceedings, Los Alamos Symposium 1989 , edited by K. S. Bedell, D. Coffey, D. E. Meltzer, D. Pines and J. R. Schrieffer (Addison-Wesley, 1990), p. 37
9. J. Orenstein, G.A. Thomas, A.J. Millis, S.L. Cooper, D.H. Rapkine, T. Timusk, L.F. Schneemeyer and J.V. Waszczak, Phys. Rev. B **42**, 6342 (1990)
10. K.F. Renk, J. Schutzmann, A. Pruckl, B. Roas, L. Schultz and G. Saemann-Ischenko, Physica B **165&166**, 1253 (1990)
11. K.F. Renk, B. Gorshunov, J. Schutzmann, A. Pruckl, B. Brunner, J. Betz, S. Orbach, N. Klein, G. Muller and H. Piel, (preprint)

12. T.L. Hylton, A. Kapitulnik, M.R. Beasley, J.P. Carini, L. Drabeck and G. Gruner, Applied Physics Letters **53**, 1343 (1988)

13. T.L. Hylton and M.R. Beasley, Physical Review B **39**, 9042 (1989)

14. D.R. Harshman, L.F. Schneemeyer, J.V. Waszczak, G. Aeppli, R.J. Cava, B. Batlogg, L.W. Rupp, E.J. Ansaldo and D.L. Williams, Physical Review B **39**, 851 (1989)

15. R. Gross, P. Chaudhari, M. Kawasaki and A. Gupta, IEEE Trans. Magn. **27**, 3227 (1991)

16. C.B. Eom, J.Z. Sun, B.M. Lairson, S.K. Streiffer, A.F. Marshall, K. Yamamoto, S.M. Anlage, J.C. Bravman, T.H. Geballe, S.S. Laderman, R.C. Taber and R.D. Jacowitz, Physica C **171**, 354 (1990)

17. S.S. Laderman, R.C. Taber, R.D. Jacowitz, J.L. Moll, C.B. Eom, T.L. Hylton, A.F. Marshall, T.H. Geballe and M.R. Beasley, Phys. Rev. B **43**, 2922 (1991)

18. T. Timusk and D.B. Tanner, in Physical Properties of High Temperature Superconductors I , edited by D. M. Ginsberg (World Scientific, 1989), p. 339

19. K. Kamaras, L.L. Herr, C.D. Porter, N. Tache, D.B. Tanner, S. Etemad, T. Venkatesan, E. Chase, A. Inam, X.D. Wu, M.S. Hegde and B. Dutta, Phys. Rev. Lett. **64**, 84 (1990)

Table I: Samples measured in this work. Values of microwave surface resistance R_s measured at 4K near 10 GHz are scaled to 10 GHz using a frequency squared dependence.

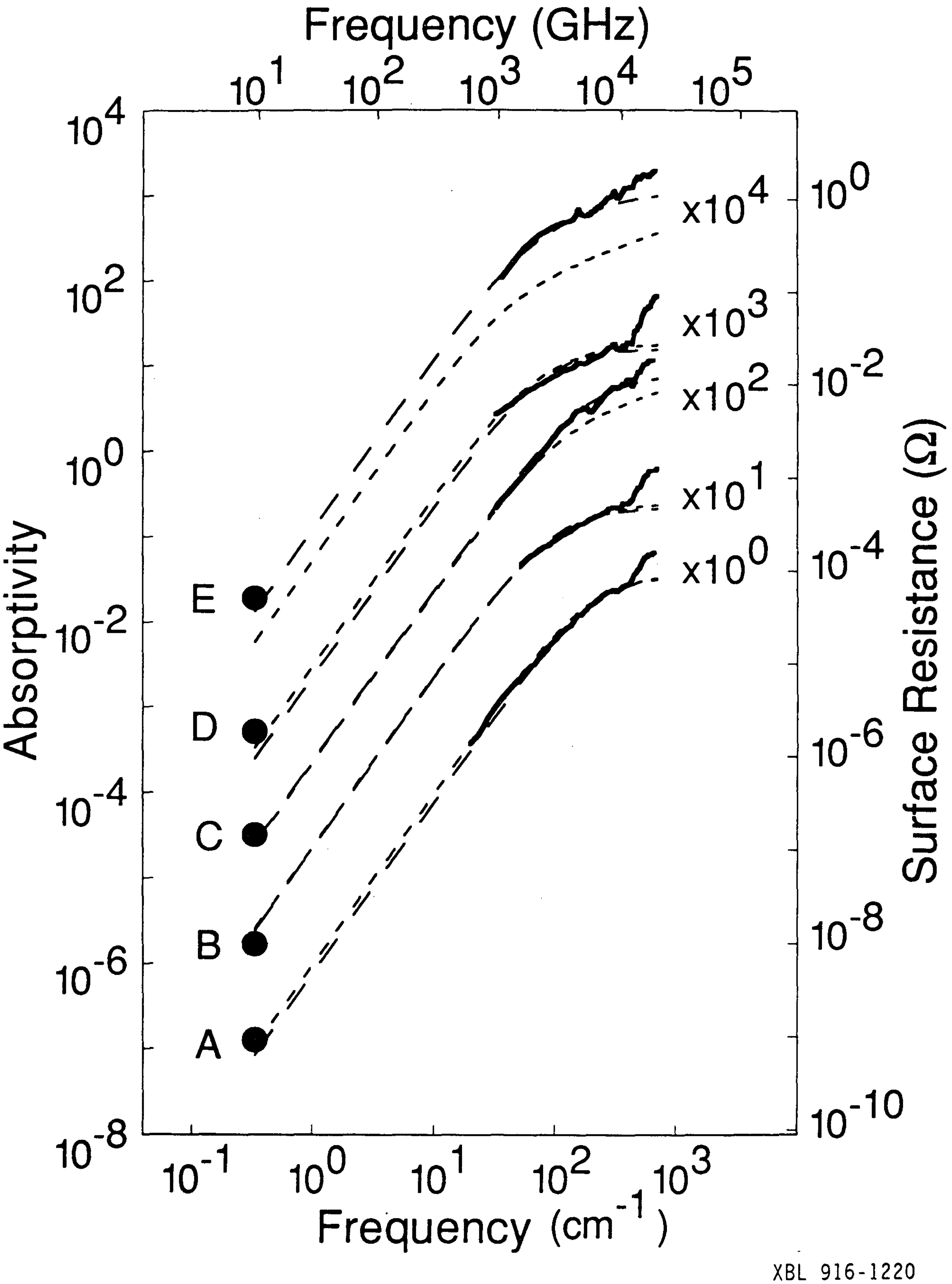
Sample	Institution	thickness (nm)	Substrate	$T_c/\delta T$ (K)	Deposition Technique	R_s ($\mu\Omega$)
A	Stanford	500	MgO	85 / 1.0	Off-Axis Sputter	12
B	Stanford	400	MgO	85 / 1.0	Off-Axis Sputter	16
C	Conductus	410	$LaAlO_3$	87 / 1.0	Off-Axis Sputter	30
D	Bellcore	500	$LaAlO_3$	92 / 0.5	Laser Ablation	48
E	Conductus	1250	$LaAlO_3$	87 / 2.0	Off-Axis Sputter	180

Table II: Parameters λ_j and ρ_J of the weakly coupled grain model computed for $\lambda_g = 140$ nm, and parameters ρ_o and $\hbar/\tau$ of the two-fluid model computed for $\lambda_{tf} = 140$ nm, as described in the text.

Sample	λ_j (nm)	ρ_J ($\mu\Omega$ cm)	ρ_o ($\mu\Omega$ cm)	$\hbar/\tau$ (cm^{-1})
A	148 ± 17	93 ± 25	86 ± 18	566 ± 210
B	175 ± 30	57 ± 18	36 ± 10	426 ± 215
C	265 ± 55	216 ± 90	34 ± 12	>280
D	160 ± 20	41 ± 9	28 ± 12	305 ± 195
E	502 ± 100	302 ± 150	16 ± 15	>7000

Figure Captions

FIG. 1. Measured submillimeter absorptivities of samples A through E at 2K (solid lines) multiplied by the indicated factors to separate the curves. Values of the microwave surface resistance measured for each sample at 4K and scaled to 10 GHz are shown as filled circles. Also shown are best fits to the weakly coupled grain model (long dashed lines) and best fits to the two-fluid model (short dashed lines). Sample E was intentionally prepared to give large microwave loss.



XBL 916-1220

Figure 1

LAWRENCE BERKELEY LABORATORY
CENTER FOR ADVANCED MATERIALS
1 CYCLOTRON ROAD
BERKELEY, CALIFORNIA 94720